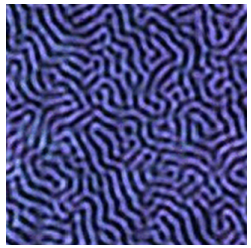
Coherent X-ray Nanovision

SRI Satellite Workshop July 13-14, 2015

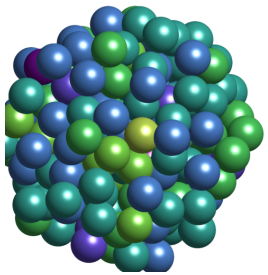
Magnets



Ionics

Photonics

Future



Oleg Shpyrko
UC San Diego

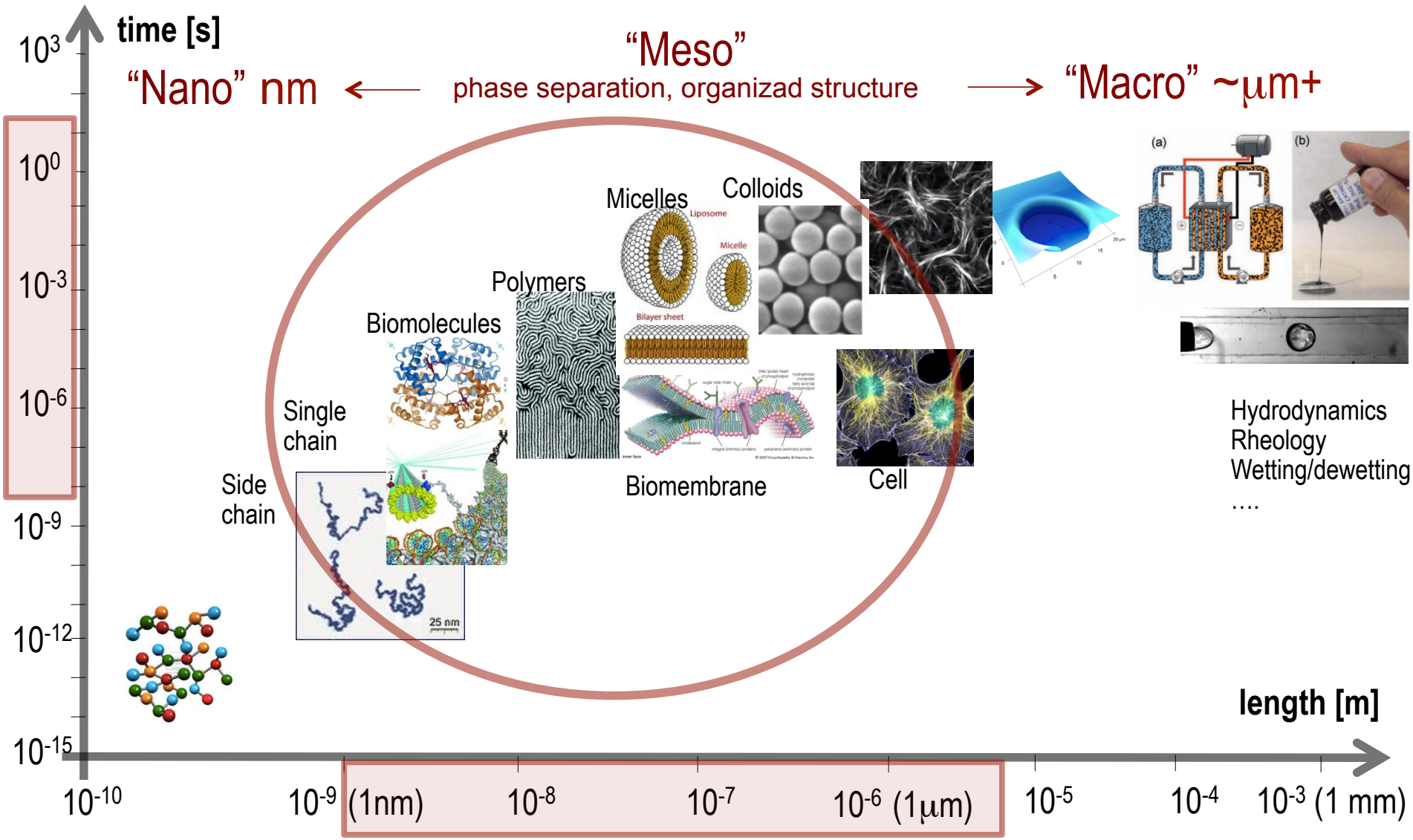
"You can observe a lot by just watching"

--Yogi Berra

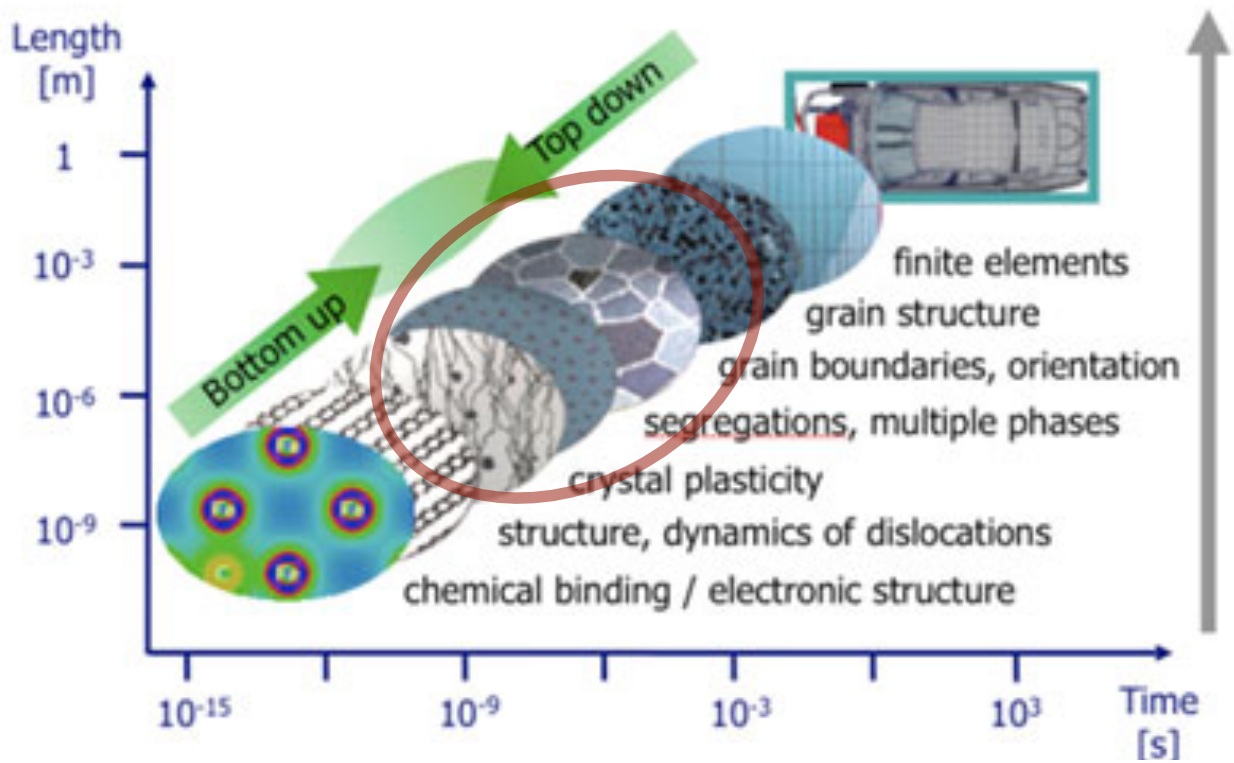
What are the ultimate limits of what we can "see" with x-rays?

How do we get there?

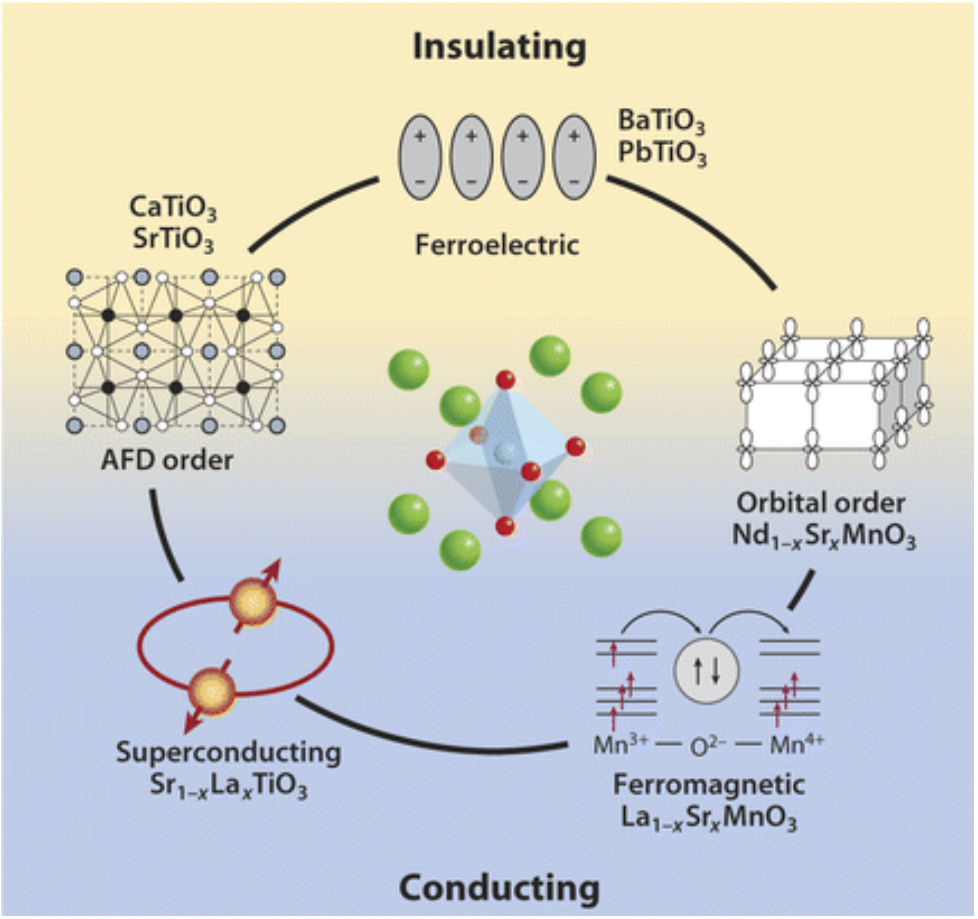
“Soft” (& “Bio”) Materials – Length and Time Scales

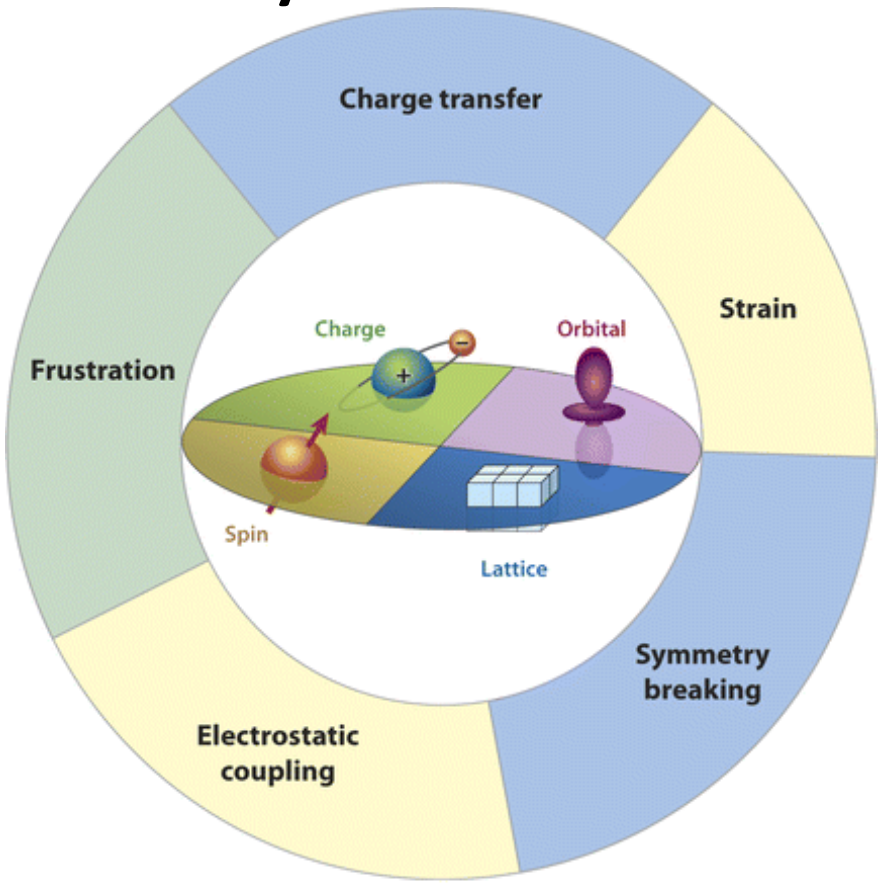


Materials Science – Length and Time Scales



Competing Ground States in Correlated Electron Systems:

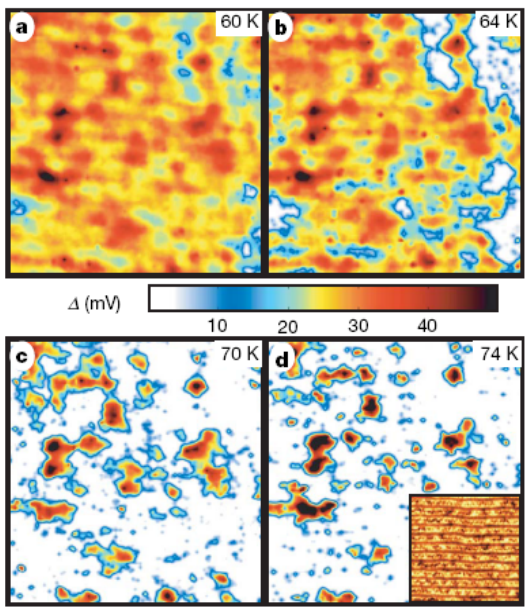




P. Zubko etal., Annu. Rev. Condens. Matter Phys. 2, 141 (2011)

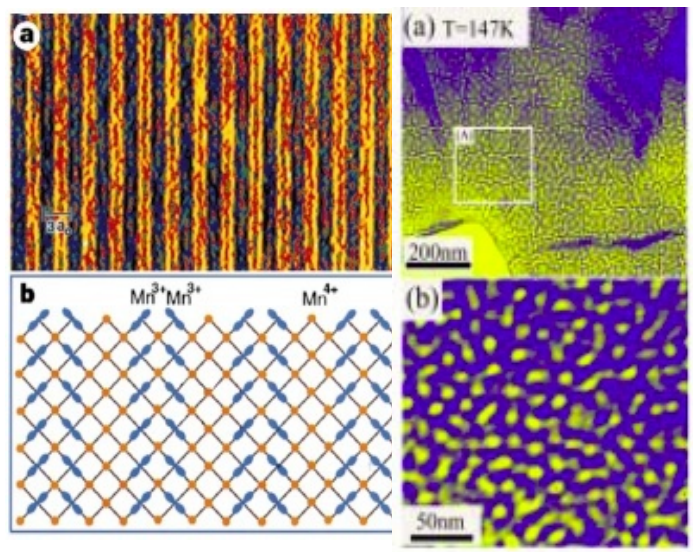
Stripes, “puddles” and domains in strongly correlated materials

High-Tc cuprates



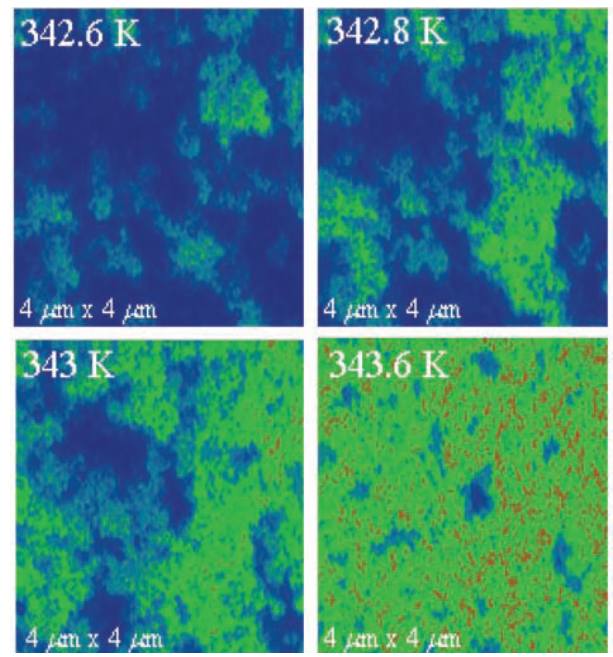
Gomes et al.,
Nature 447, 569 (2007)
SC Gap in BSCCO
(Yazdani Group, Princeton)

CMR manganites

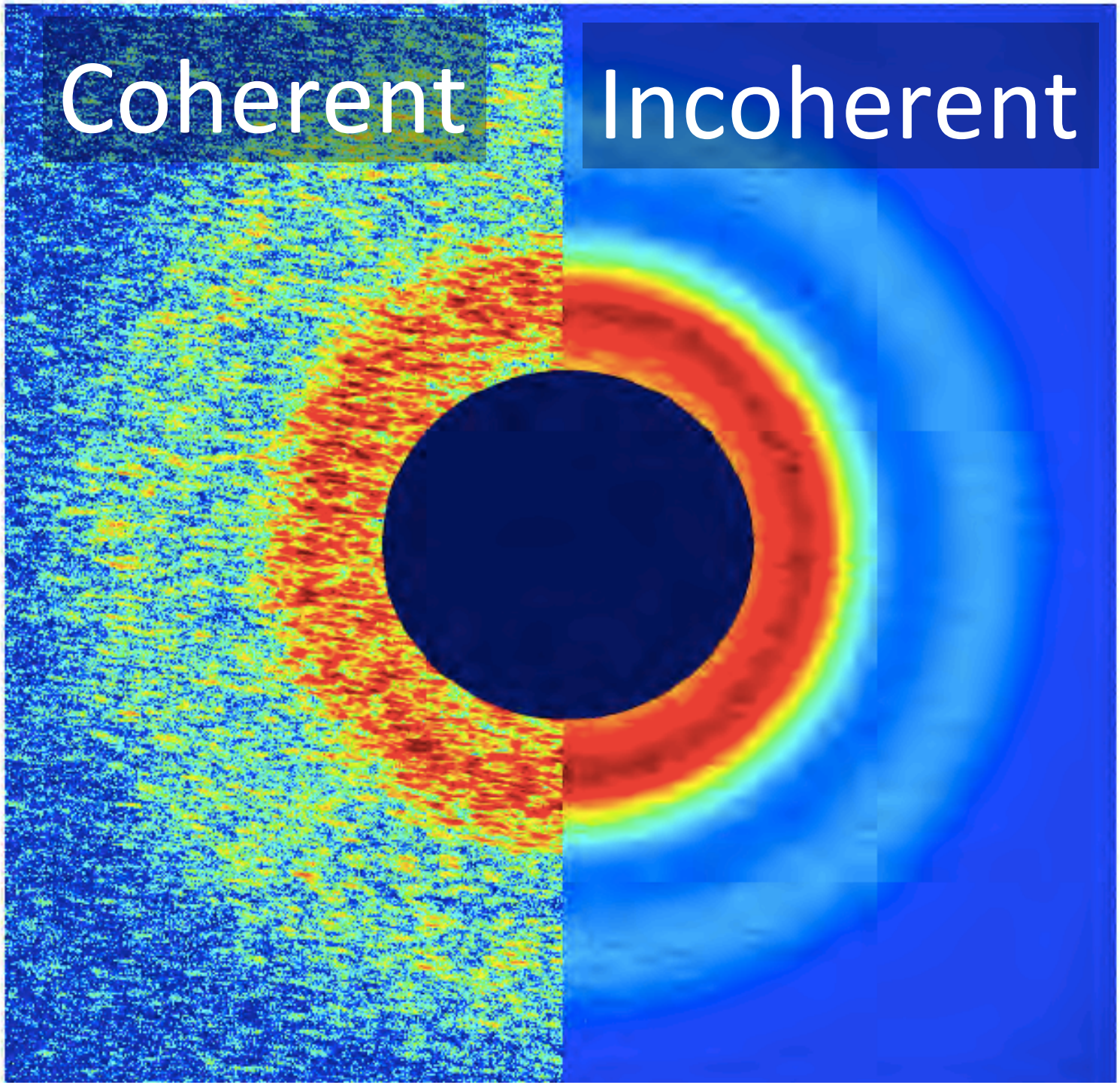


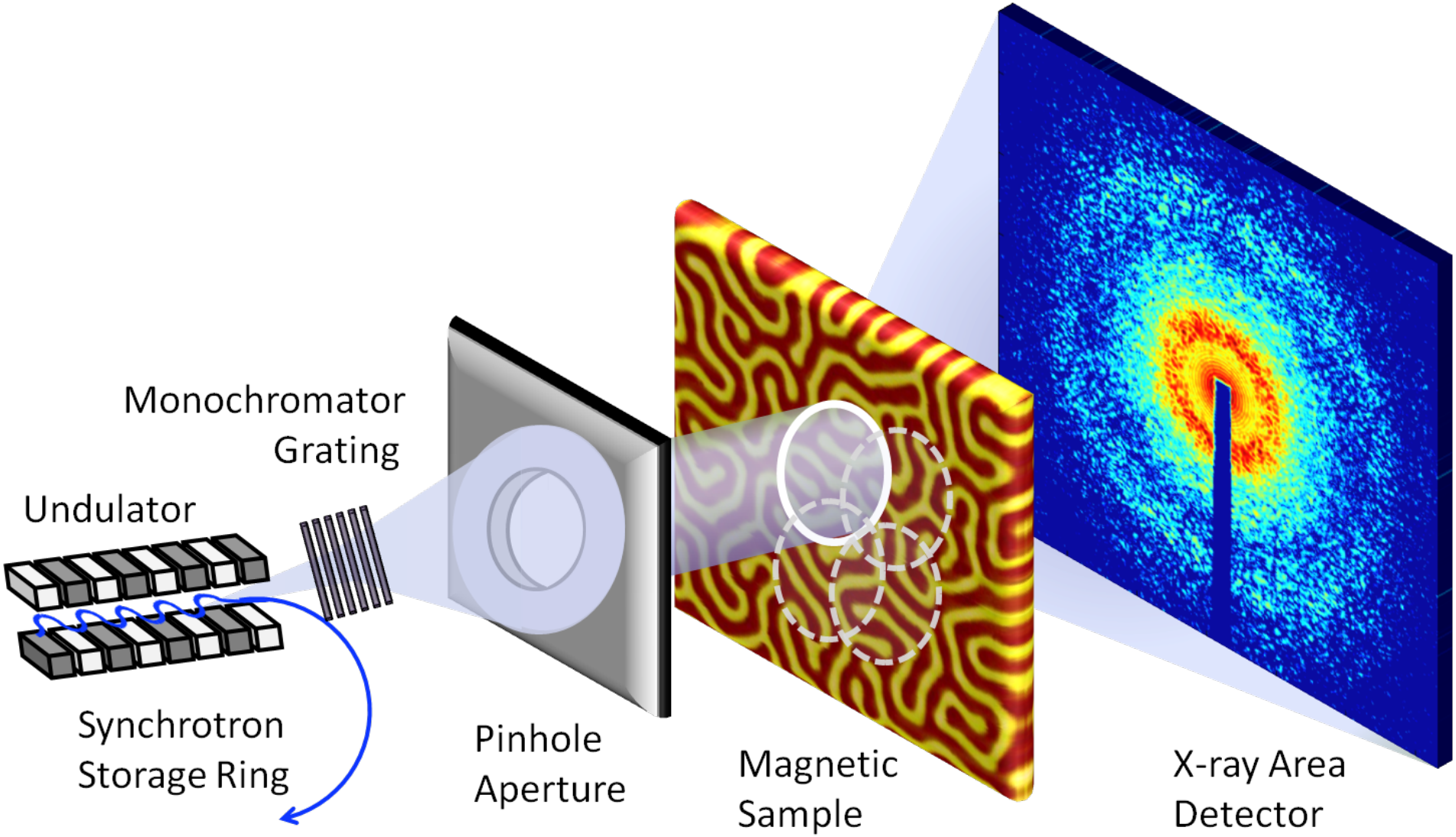
S. Mori et al., *Nature* **392,** 473 (1998)
M. Uehara et al., *Nature* **399,** 560 (1999)

Metal-Insulator
Transition In VO2



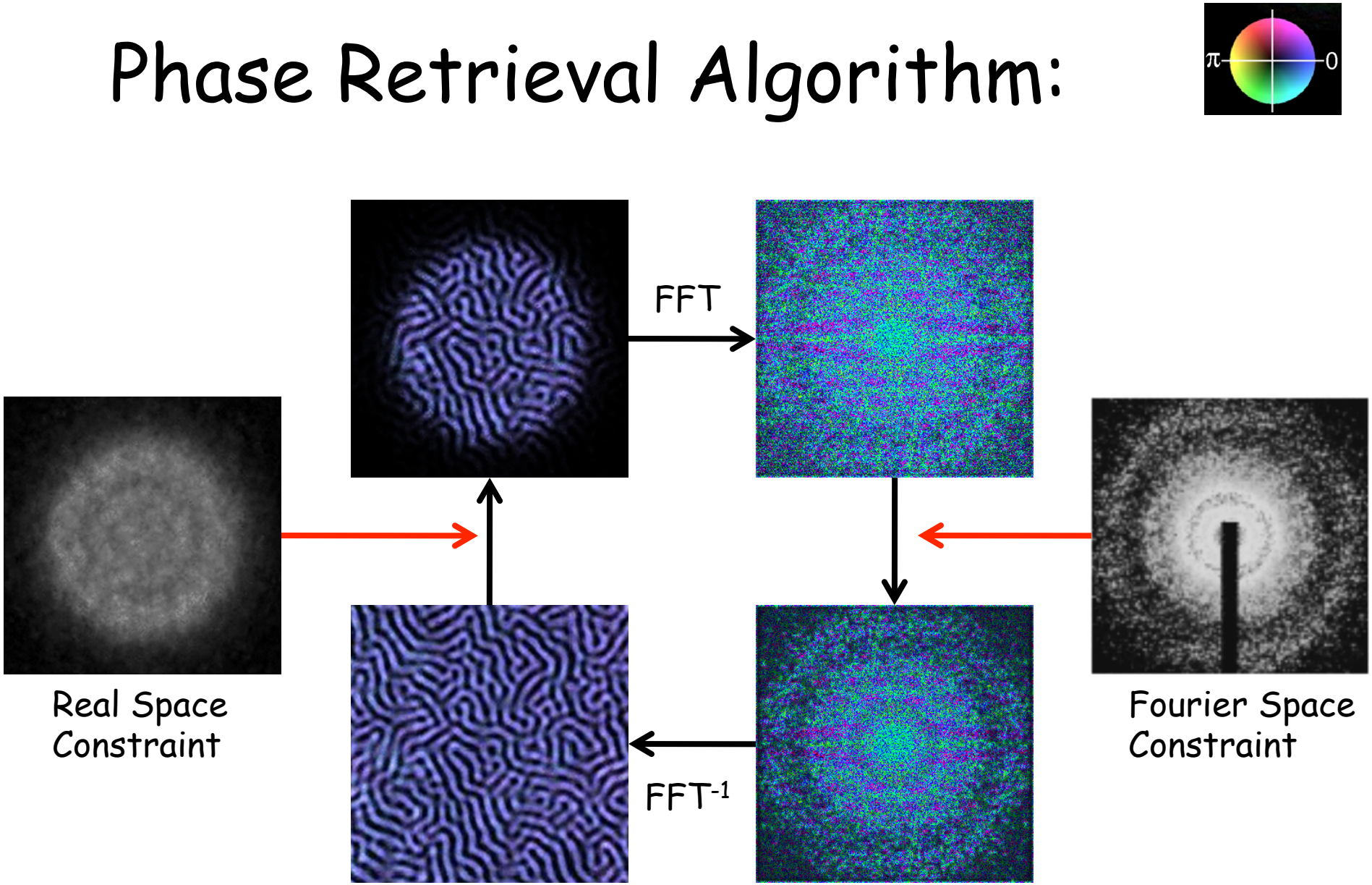
Qazilbash et al.,
Science 318, 1750 (2007)
Metal-Insulator Transition in VO2


Coherent
Incoherent



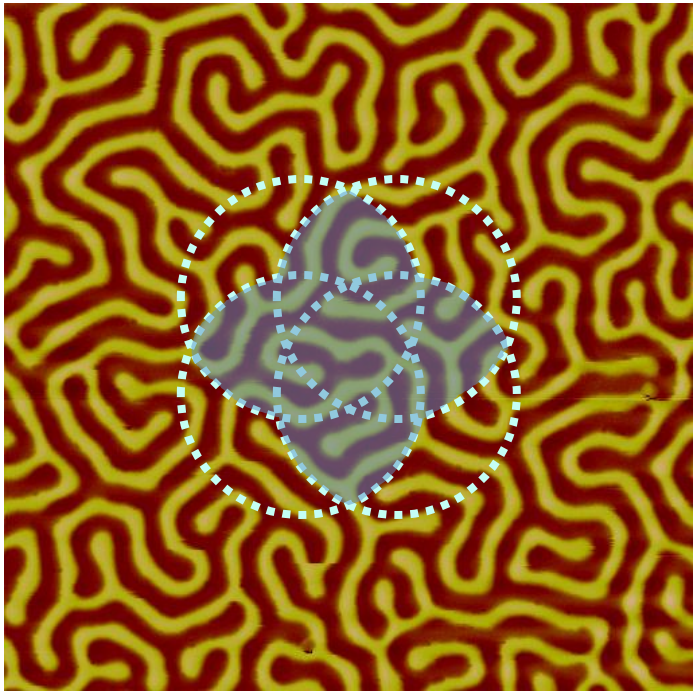
A. Tripathi, O.S. et al., Proc Natl Acad Sci USA 108, 13393 (2011)

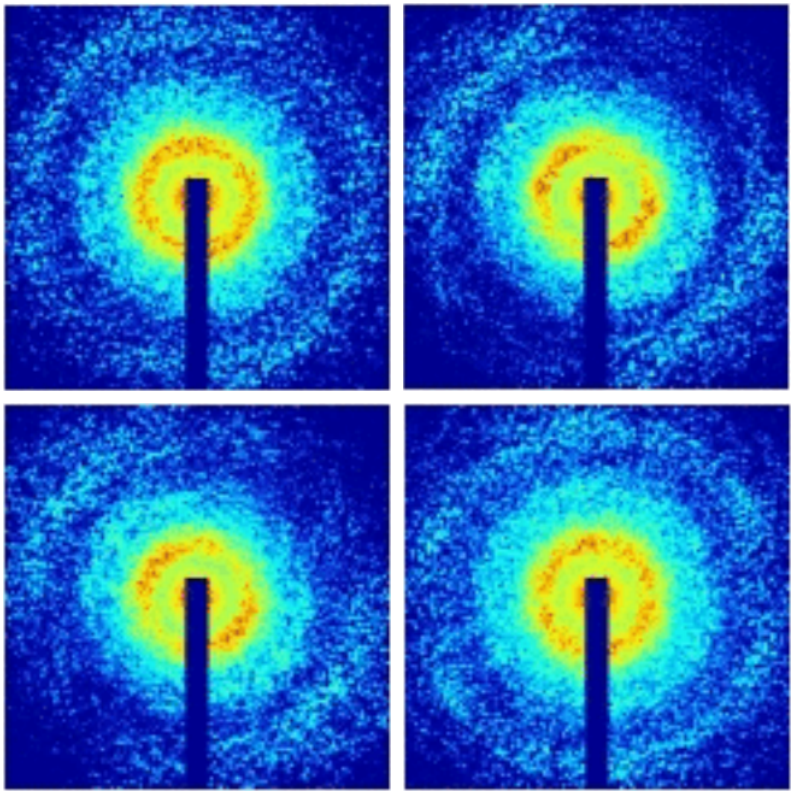
Phase Retrieval Algorithm:



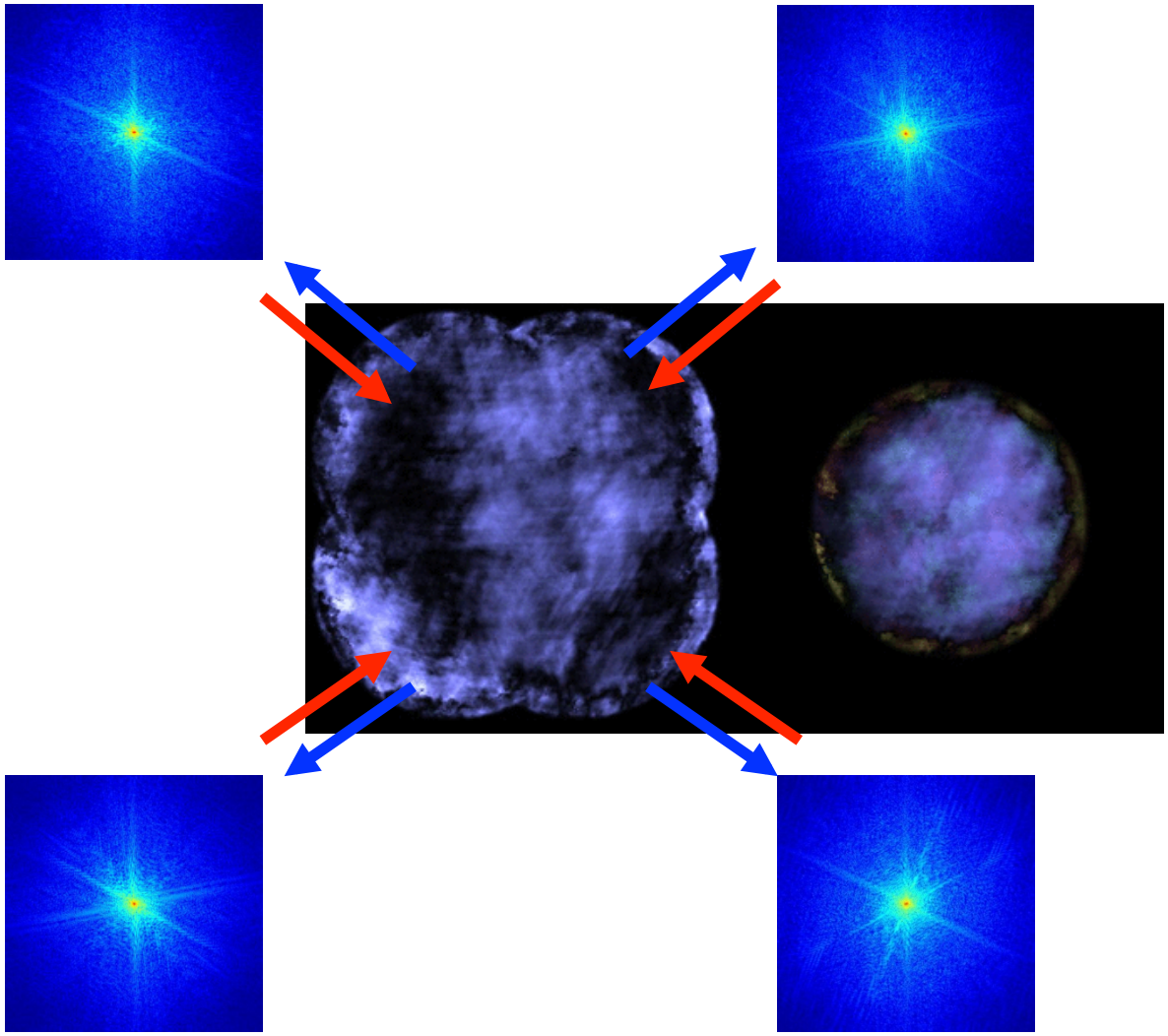
A. Tripathi, O.S. et al., Proc Natl Acad Sci USA 108, 13393 (2011)

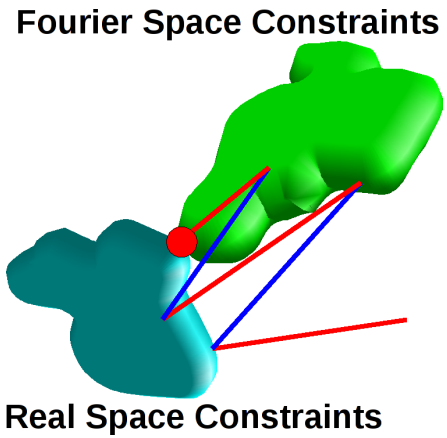
Ptychography





A. Tripathi, O.S. et al., Proc Natl Acad Sci USA 108, 13393 (2011)

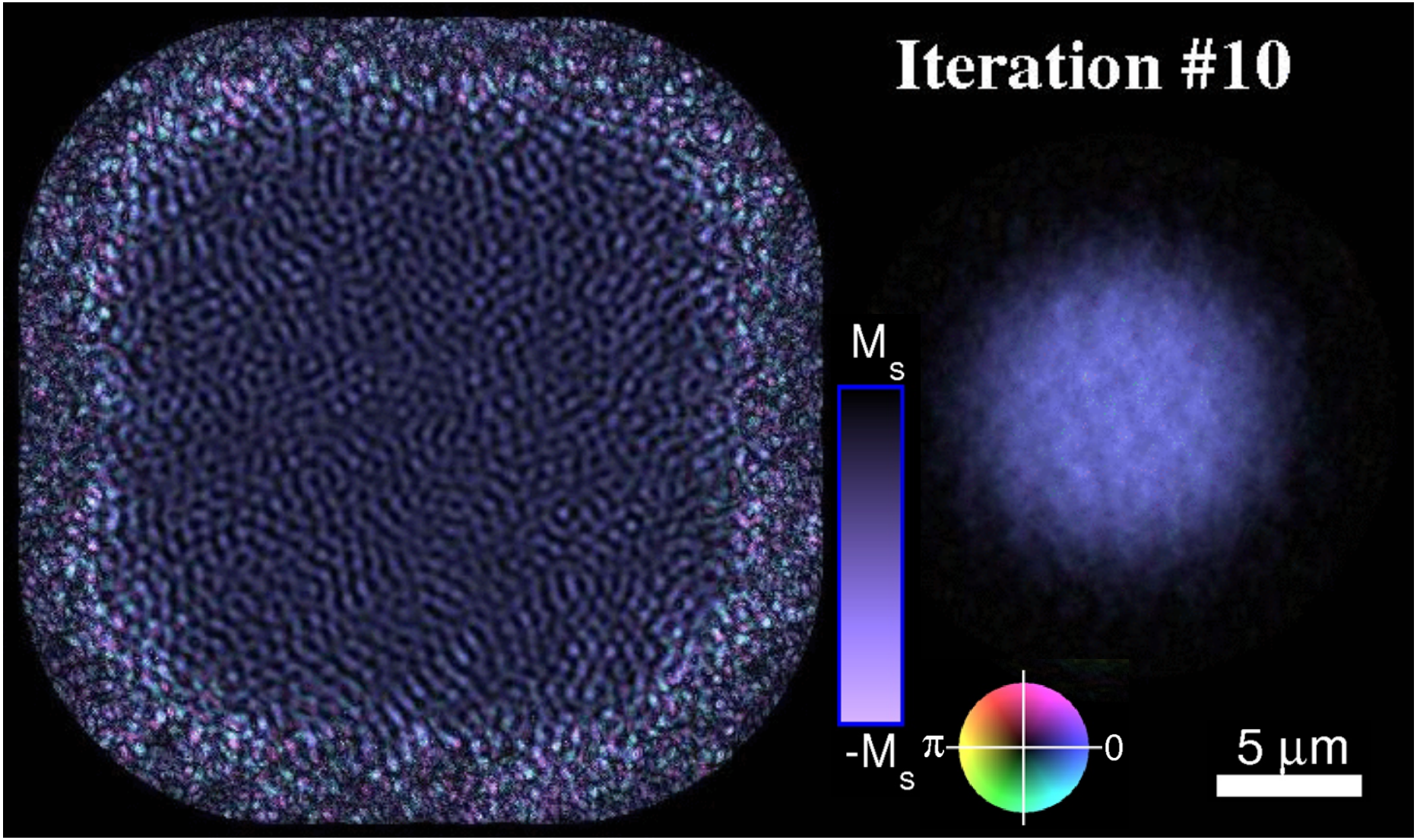




Real Space Constraint & FFT

Fourier Constraint & FFT^{-1}

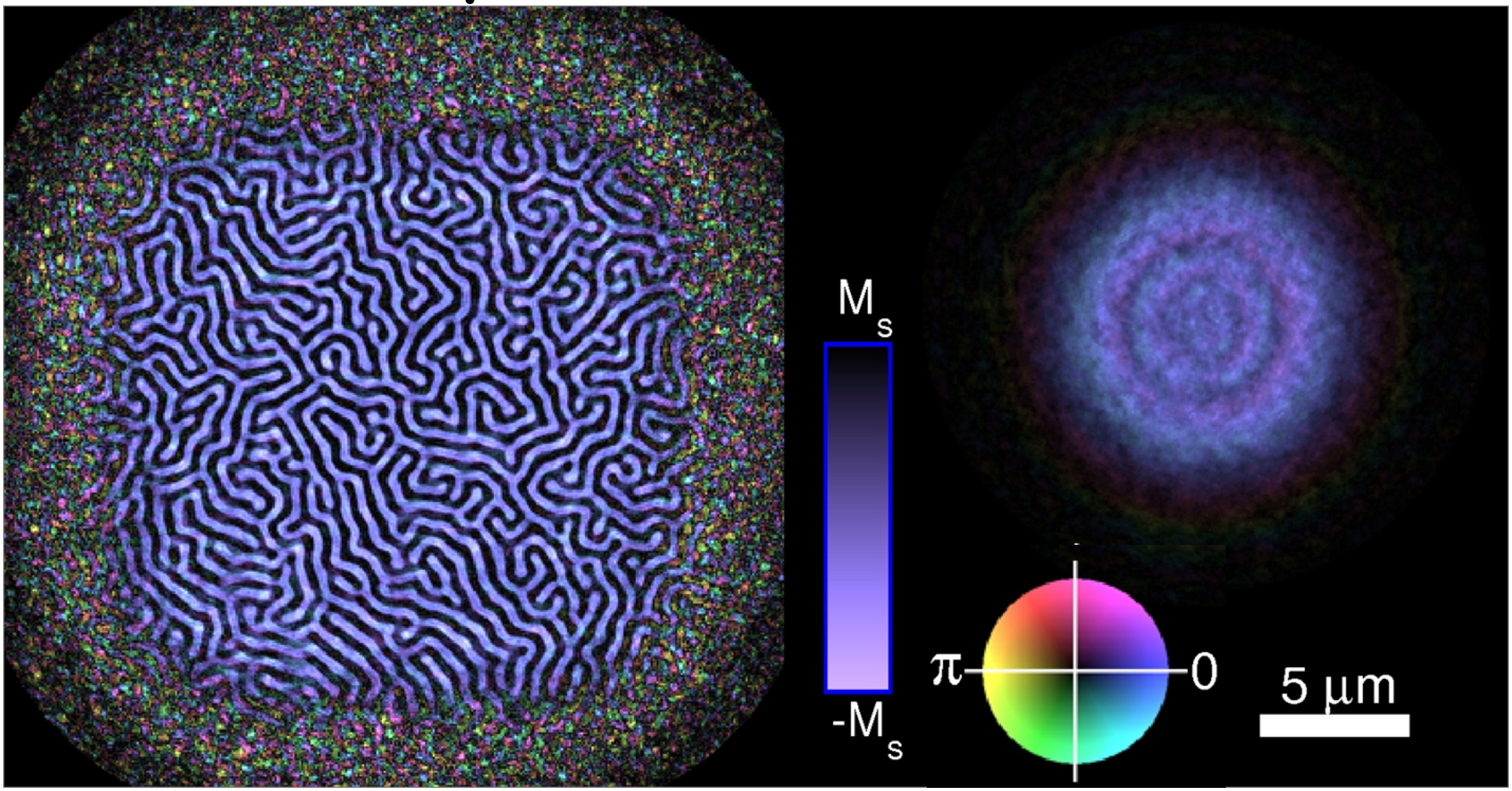
Real Space Reconstruction



Magnetic structure
(exit wave)

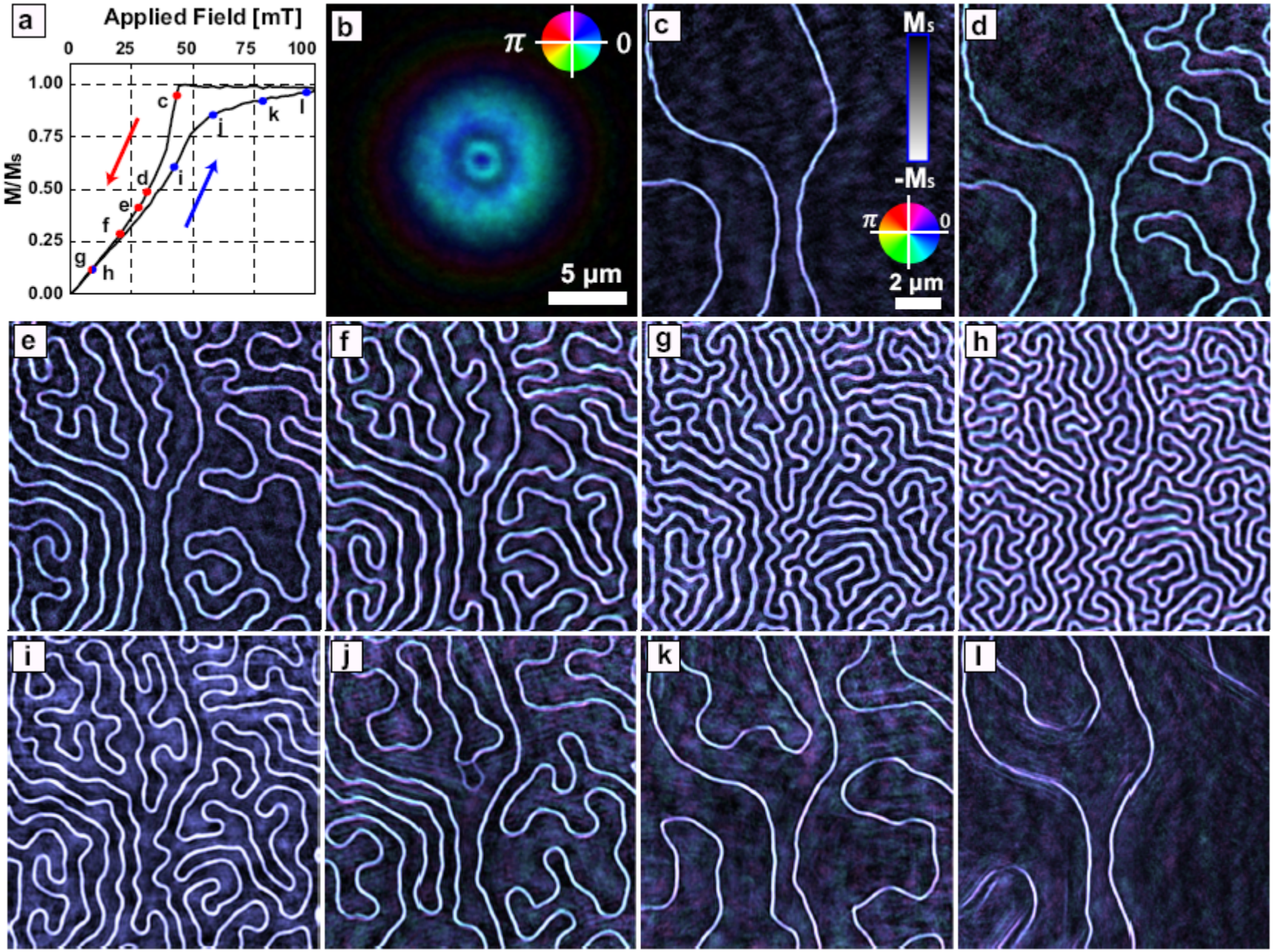
Illumination Function

Real Space Reconstruction



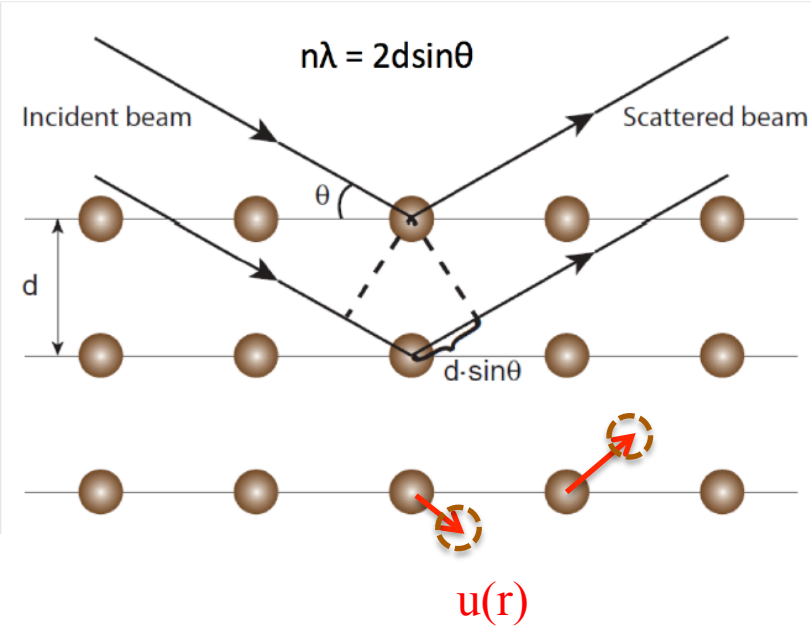
Magnetic structure
(exit wave)

Illumination Function



A. Tripathi, O.S. et al., Proc Natl Acad Sci USA 108, 13393 (2011)

Bragg Scattering Geometry: Displacement as Scattering Phase

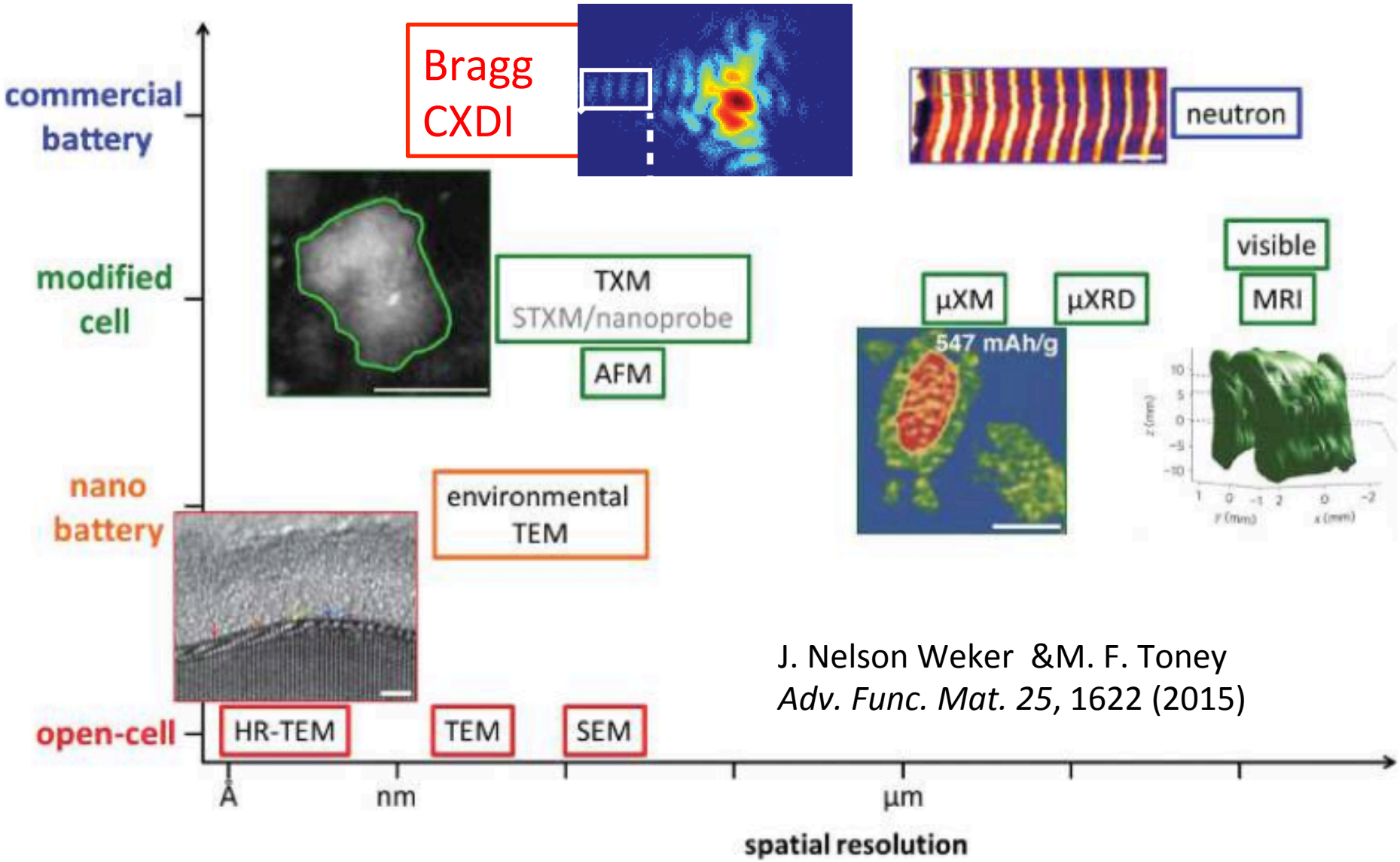


$$\rho(r) = \sum_{n=1}^{N} \sum_{j=1}^{S} \rho_{nj}(r - (R_n + r_j) - u(R_{nj}))$$

$$A(q) = F(q) \int \rho_\infty(r) S(r) e^{-iq \cdot r} dr$$

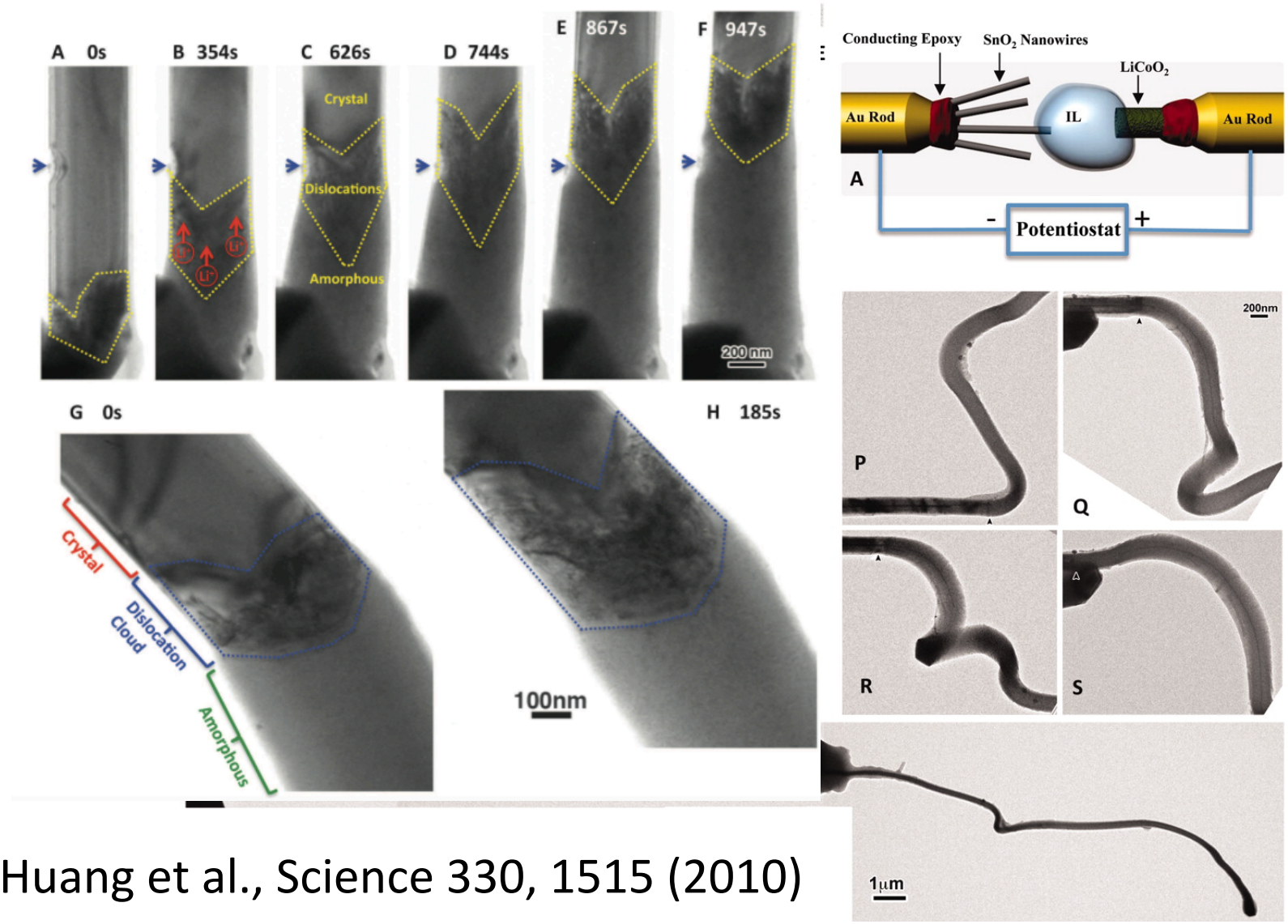
$$S(r) = s(r) e^{-iq \cdot u(r)}$$

In-operando techniques



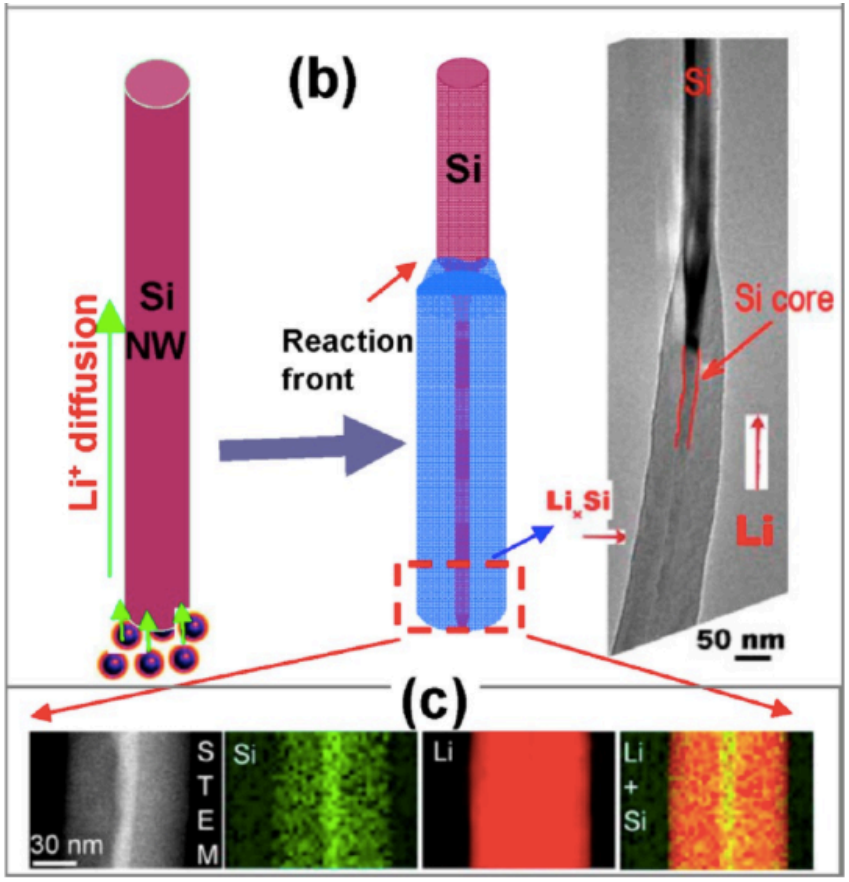
J. Nelson Weker &M. F. Toney
Adv. Func. Mat. 25, 1622 (2015)

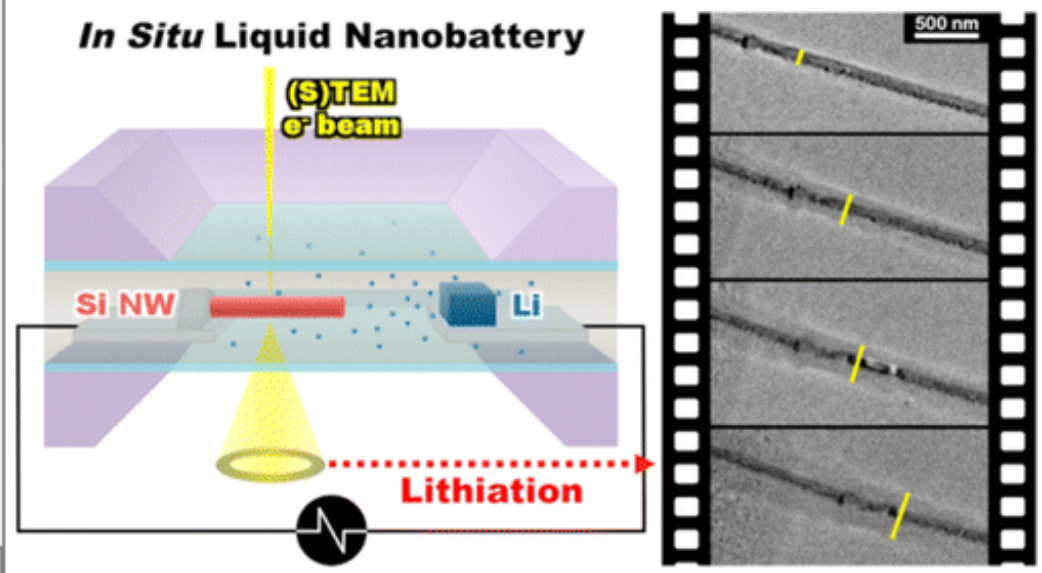
TEM in-operando imaging of ion diffusion in SnO nanowires



Huang et al., Science 330, 1515 (2010)

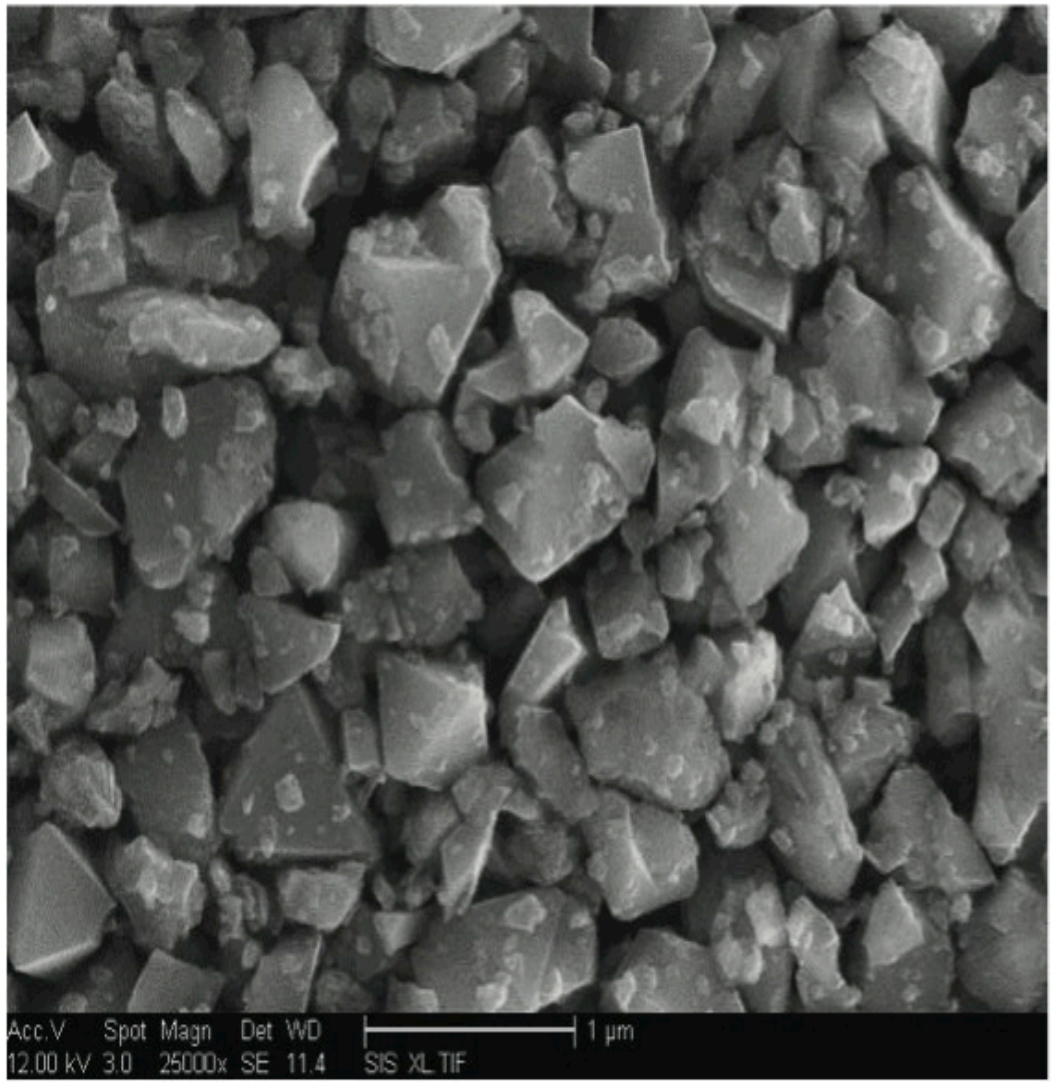
TEM in-operando imaging of ion diffusion in Si nanowires



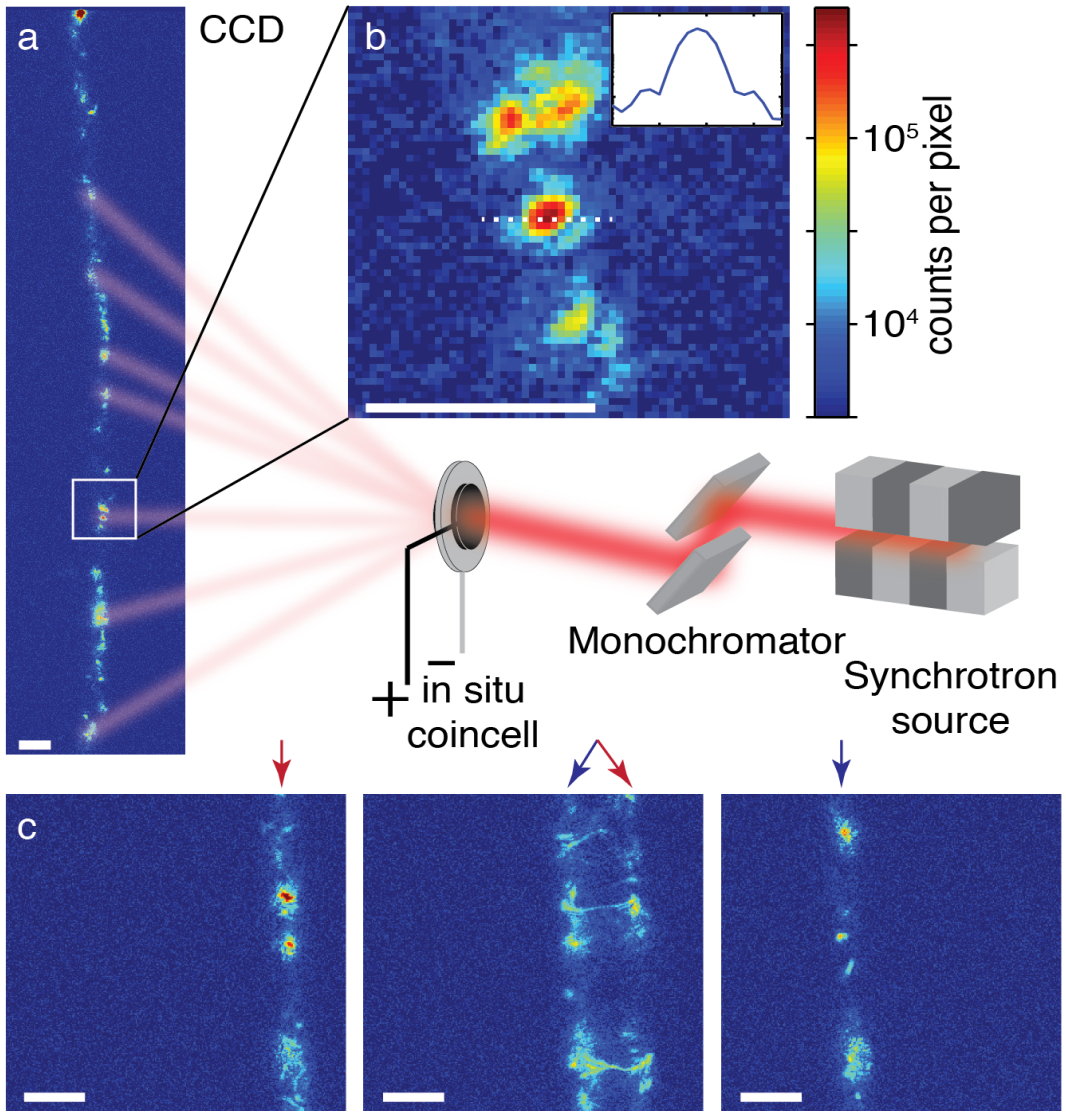


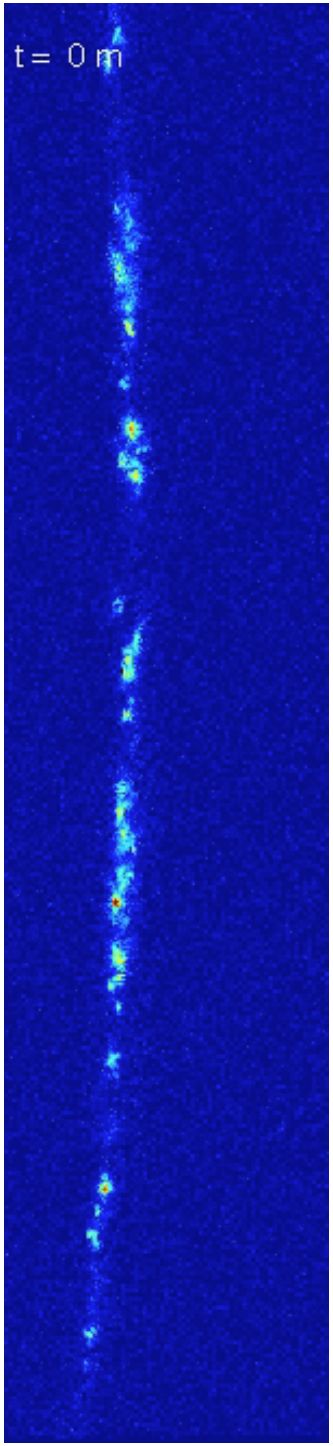
Gu et al., Nano Lett., 13, 6106 (2013)

$LiNi_{0.5}Mn_{1.5}O_4$ disordered spinel 400-700 nm particles



In-Operando CXDI of Li diffusion in $LiNi_{0.5}Mn_{1.5}O_4$





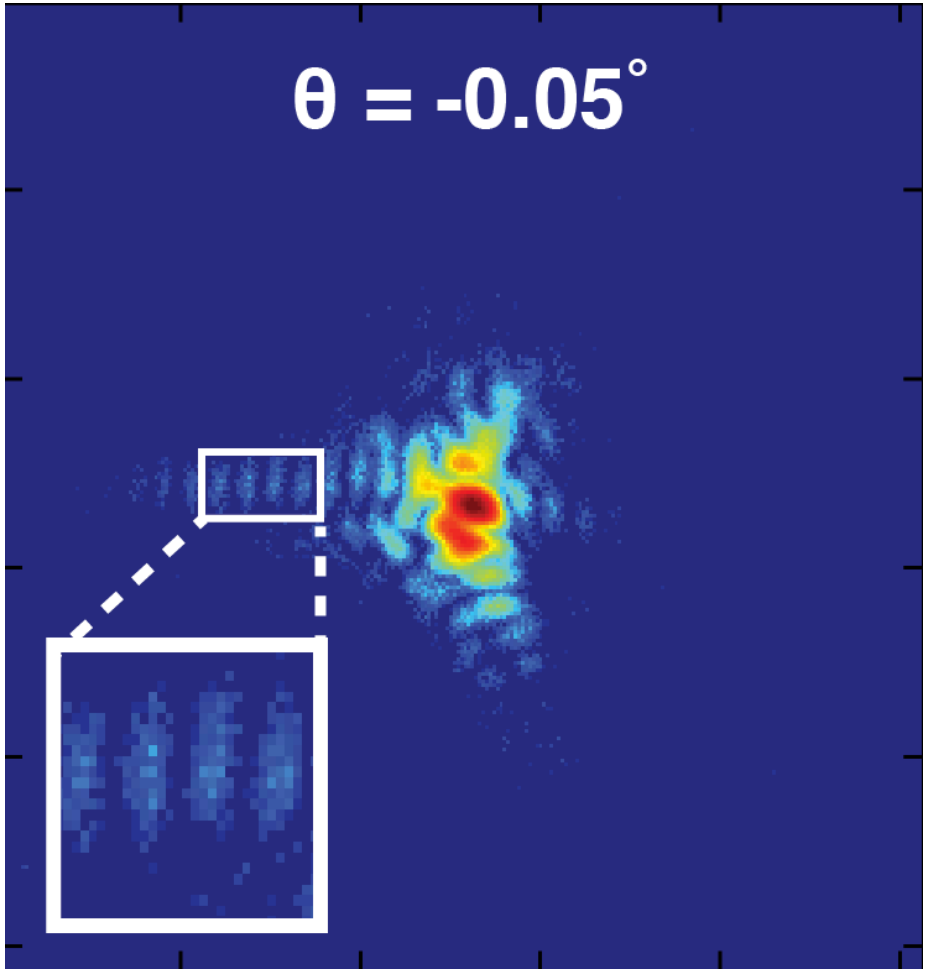
$LiNi_{0.5}Mn_{1.5}O_4$

- Full charge and full discharge in 10 hours
- 100 s per image, 600 images
- Single nano particle transformations

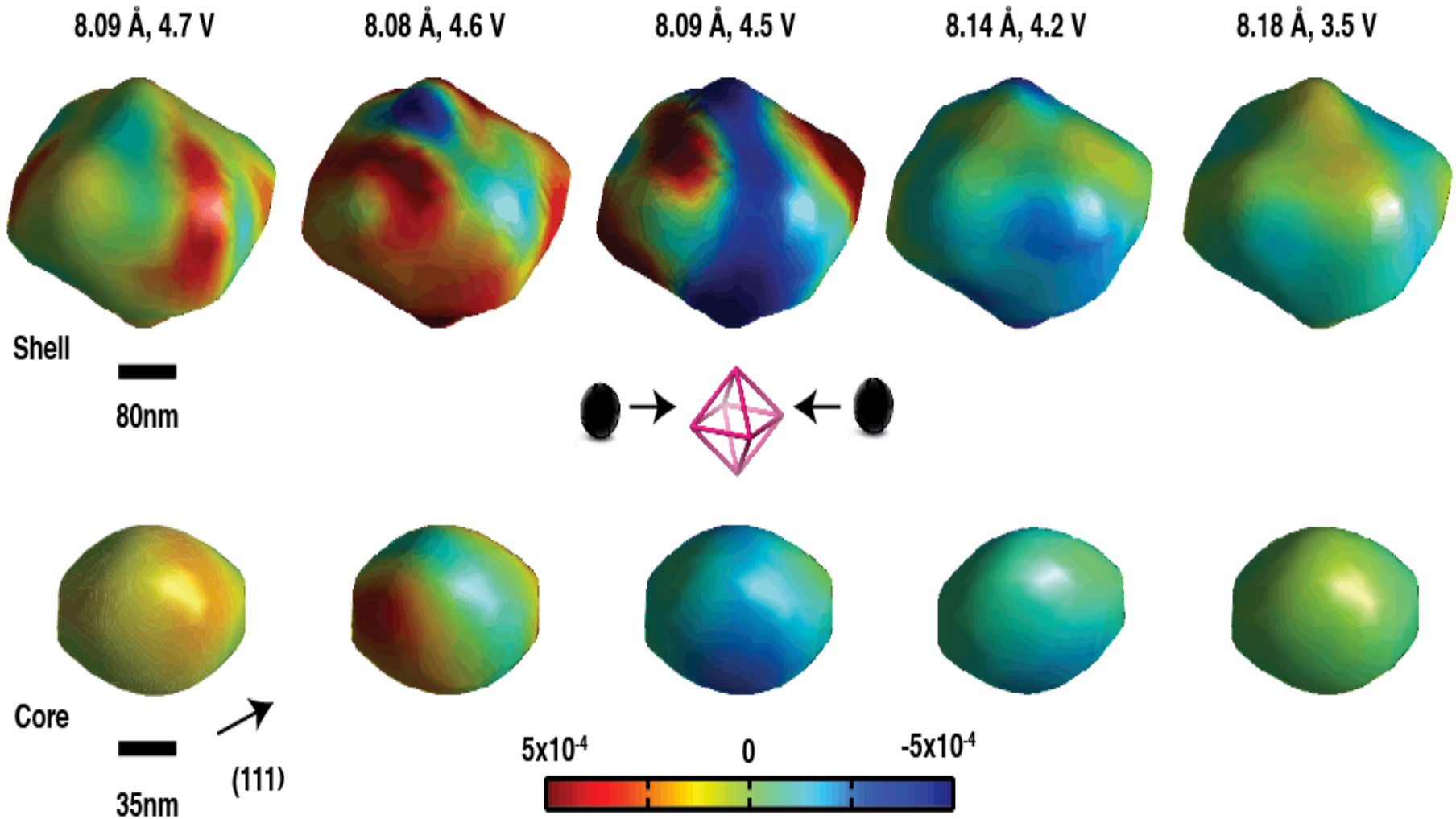
A. Singer, OS et al., Nano Lett. 14, 5295 (2014)

Phase retrieval

- Need to oversample diffraction pattern
- Coherence length > particle size
- Typically start with many (10) sets of random phases and average results

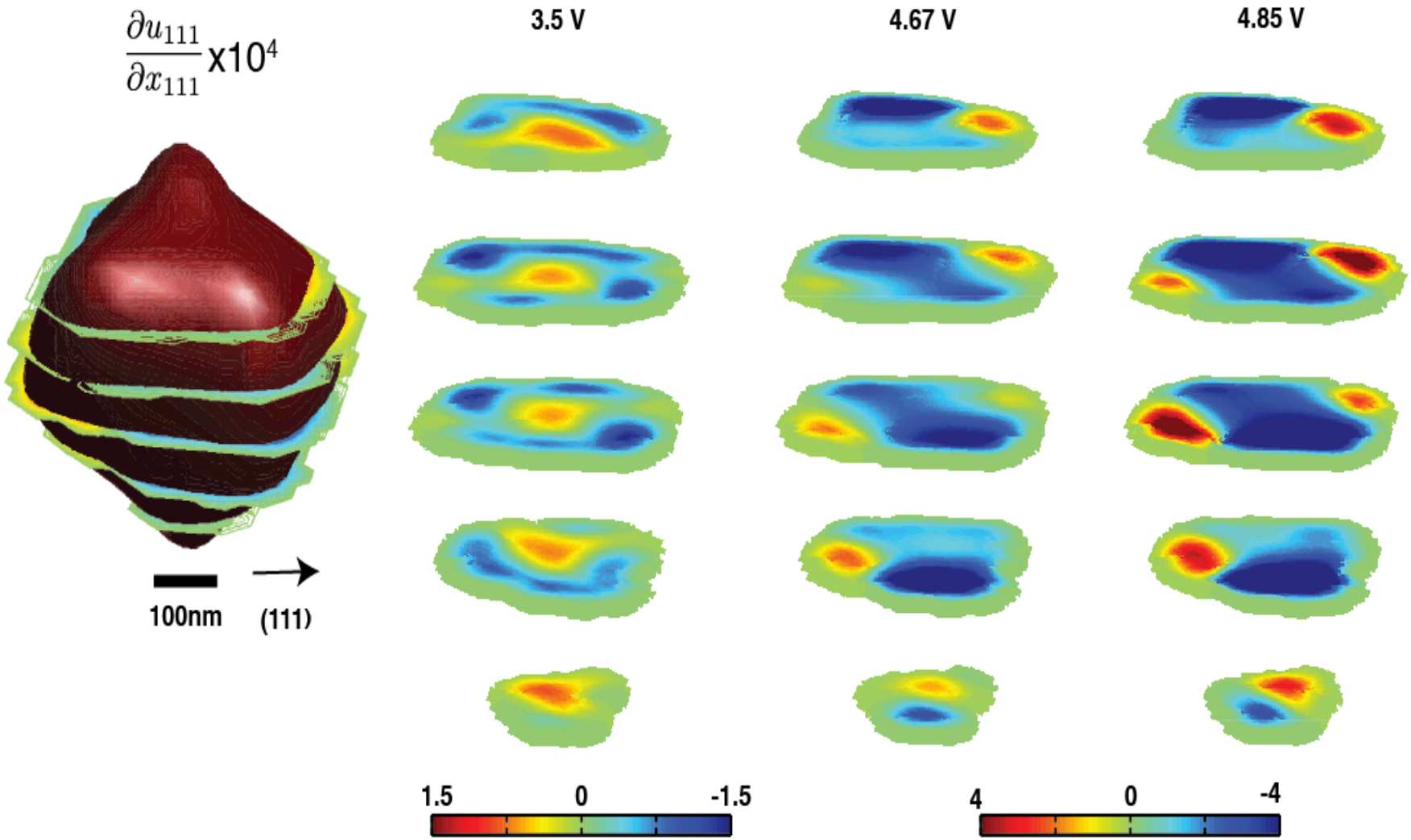


3D-Mapping of Li distribution:



A. Ulvestad, OS et al., *Nano Lett. 14, 5123 (*2014)

Strain stripe formation



A. Ulvestad, OS et al., *Nano Lett. 14, 5123 (*2014)

Strain Coherency

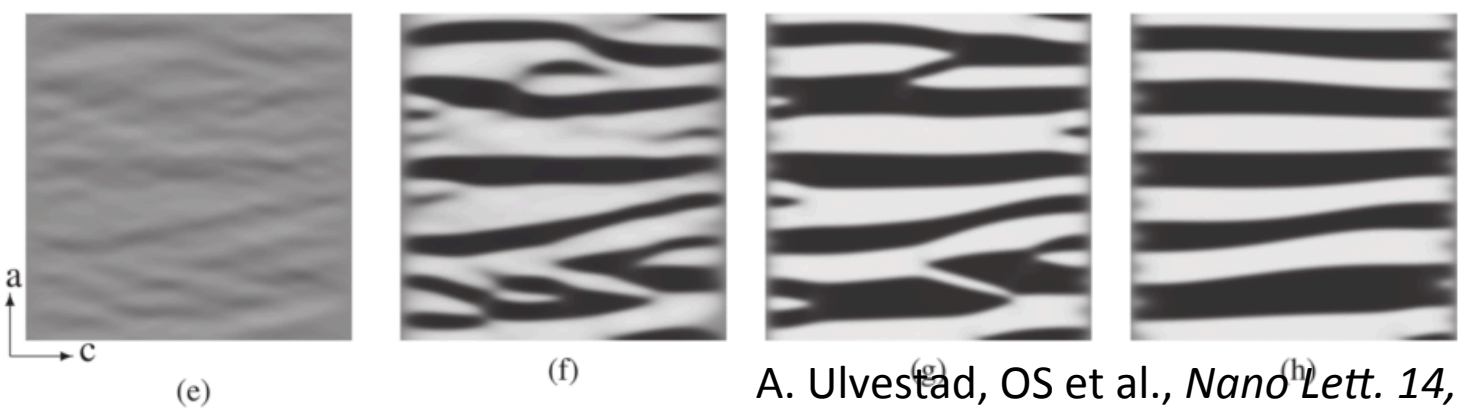
Energy = Thermodynamic contribution + gradient energy + elastic energy

$$F = N_V \int (f_0(c) + \kappa(\nabla c)^2 + \frac{1}{2}\sigma_{ij}\epsilon_{ij})\, dV$$

Thermodynamic contribution = enthalpy + entropy

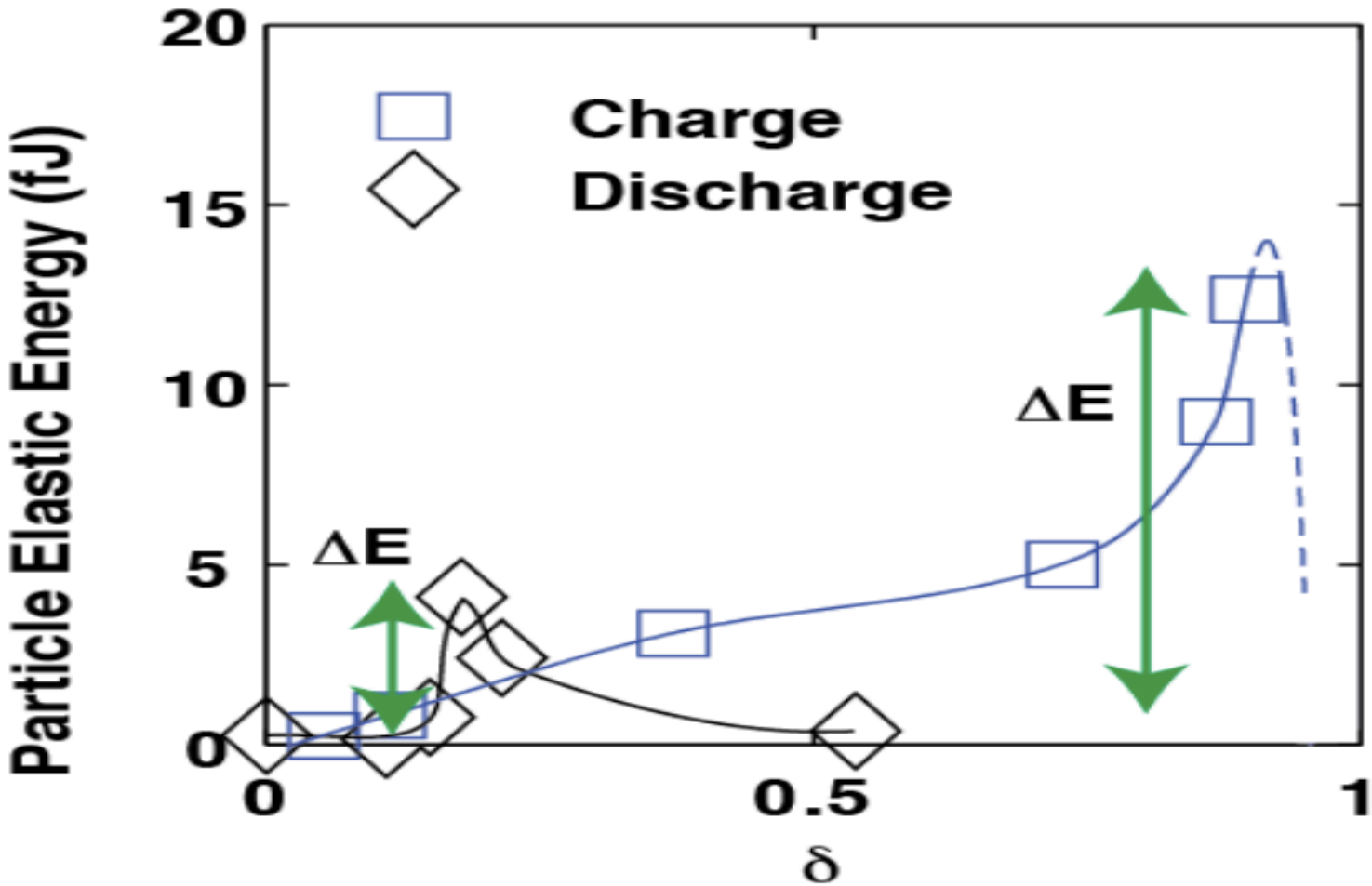
$$f_0(c) = \Omega c(1-c) + kT(c\log c + (1-c)\log(1-c))$$

Initially homogenous state decomposes into stripes (Bazant 2012)

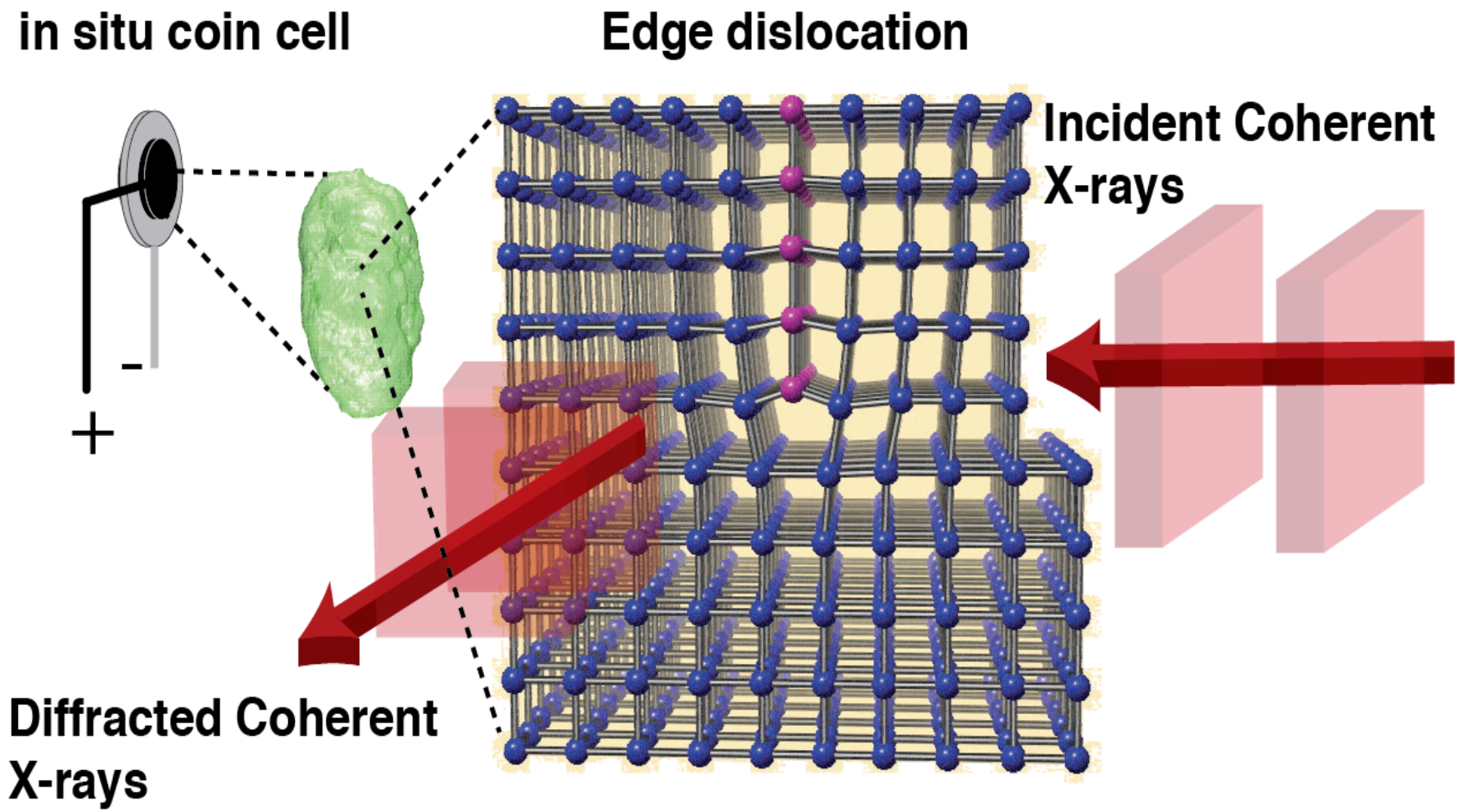


A. Ulvestad, OS et al., *Nano Lett. 14, 5123* (2014)

Elastic Energy



Can we image a single defect (e.g. dislocation)?



A. Ulvestad, et al., Science 348 (6241), 1344 (2015)

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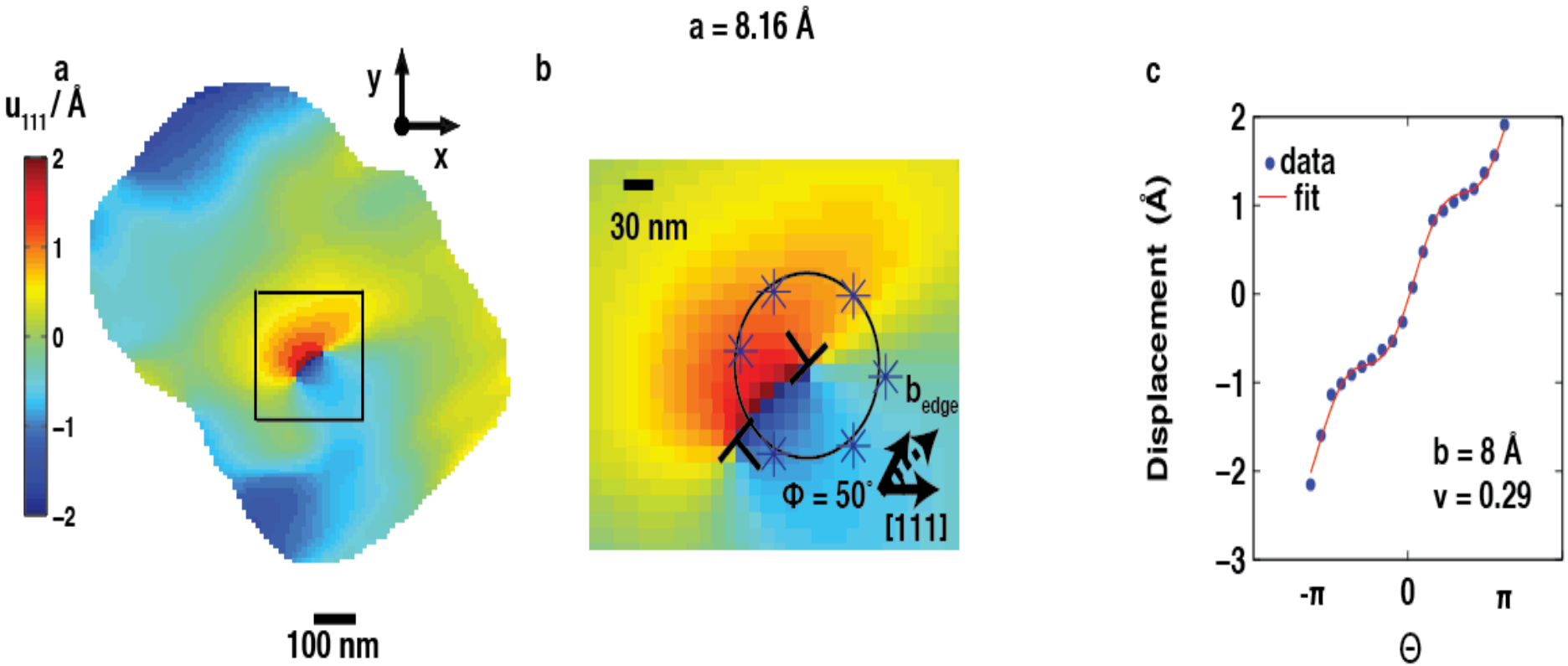

A. Ulvestad, et al., Science 348 (6241), 1344 (2015)

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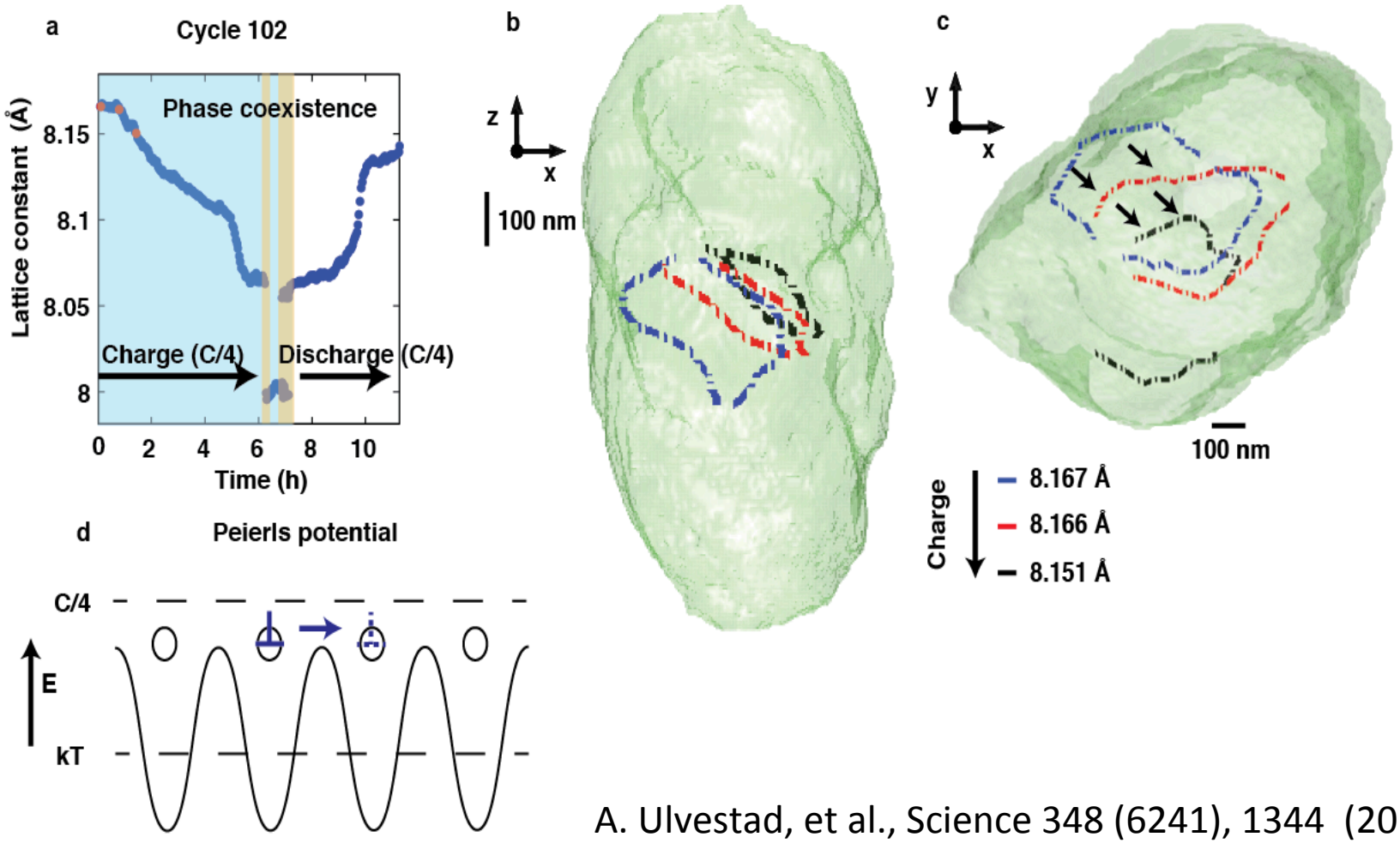
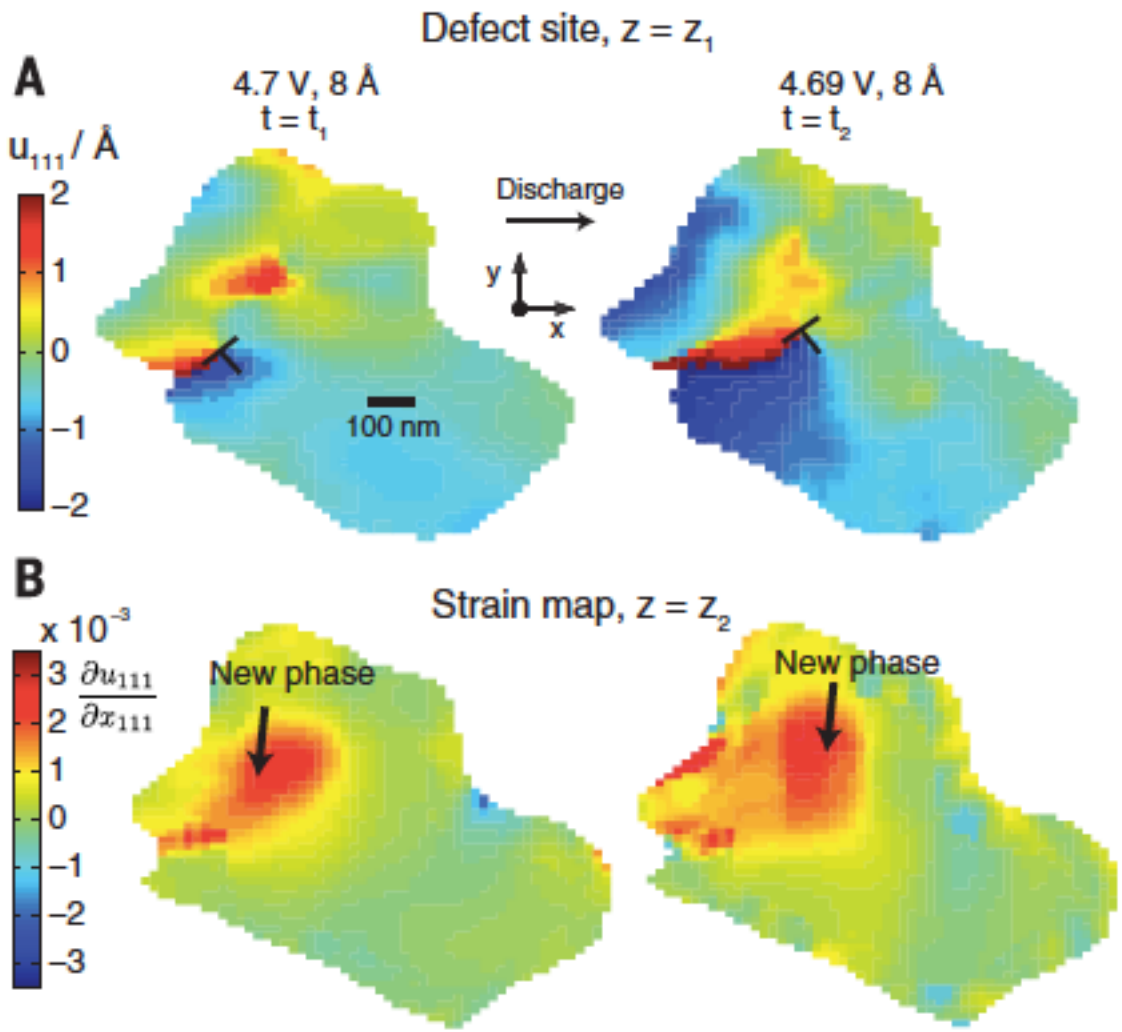


A. Ulvestad, et al., Science 348 (6241), 1344 (2015)

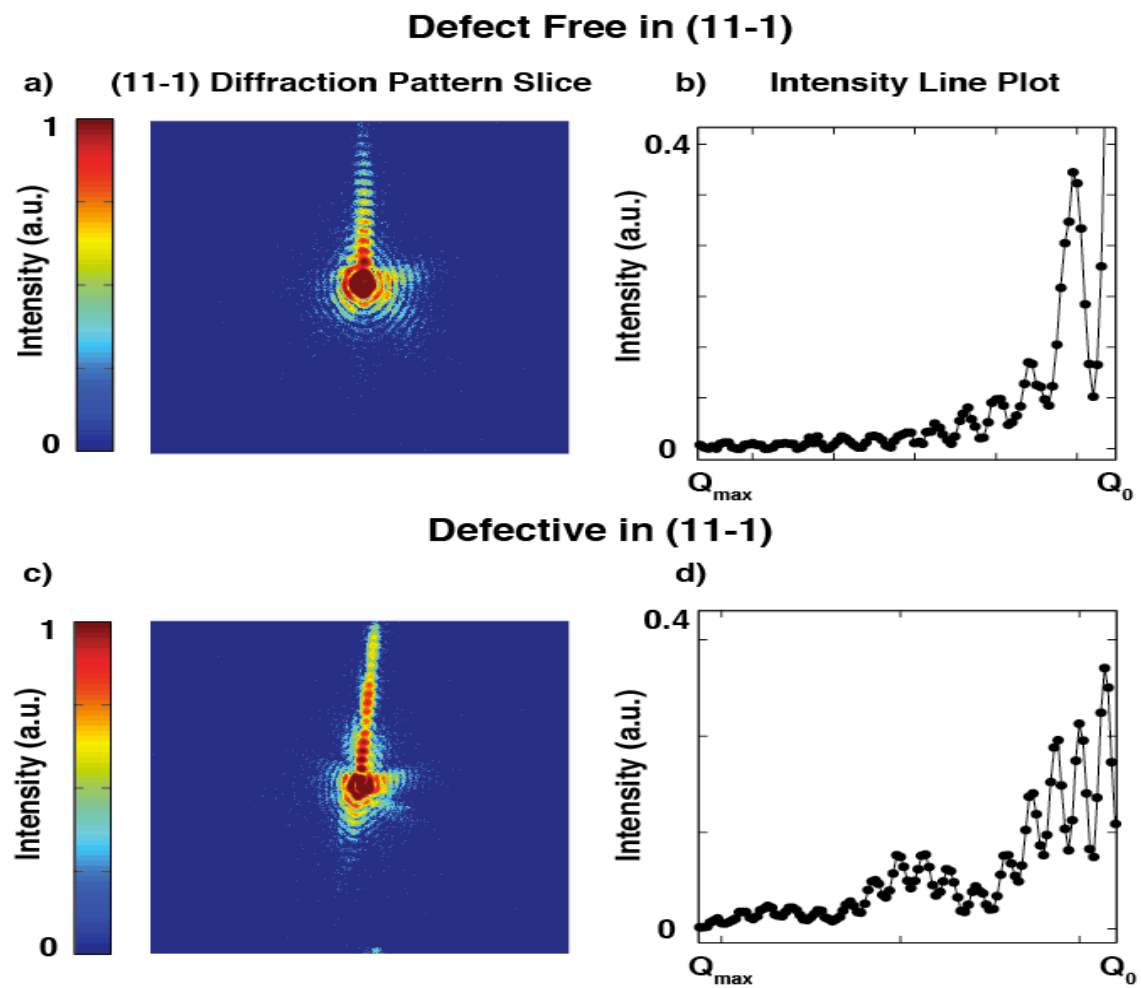
Defects as nucleation centers for phase transitions



A. Ulvestad, et al., Science 348 (6241), 1344 (2015)

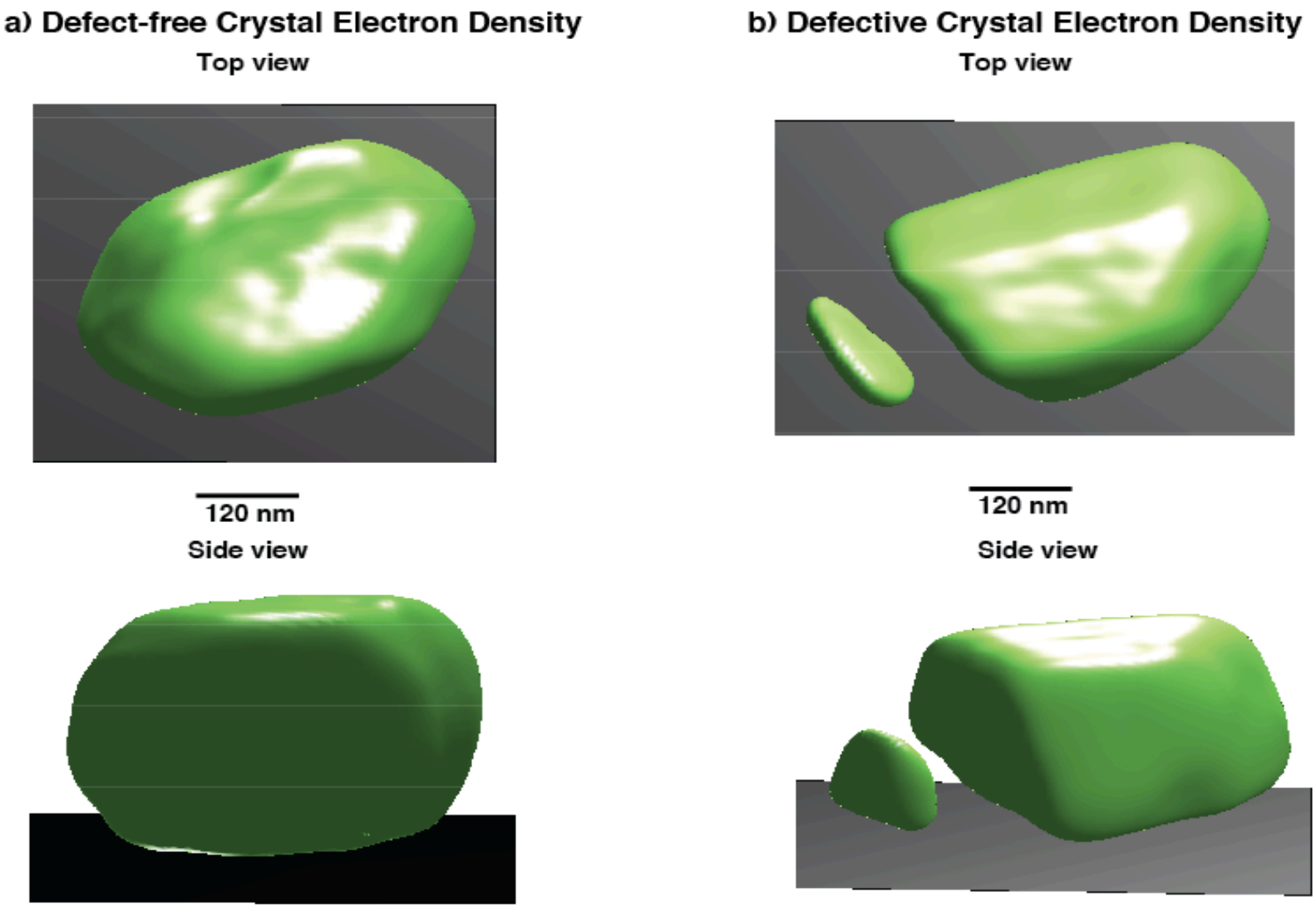
Defects in Gold Nanocrystals*

- Defect free crystal has standard fringe profile
- Defective crystal shows additional enveloping of intensity

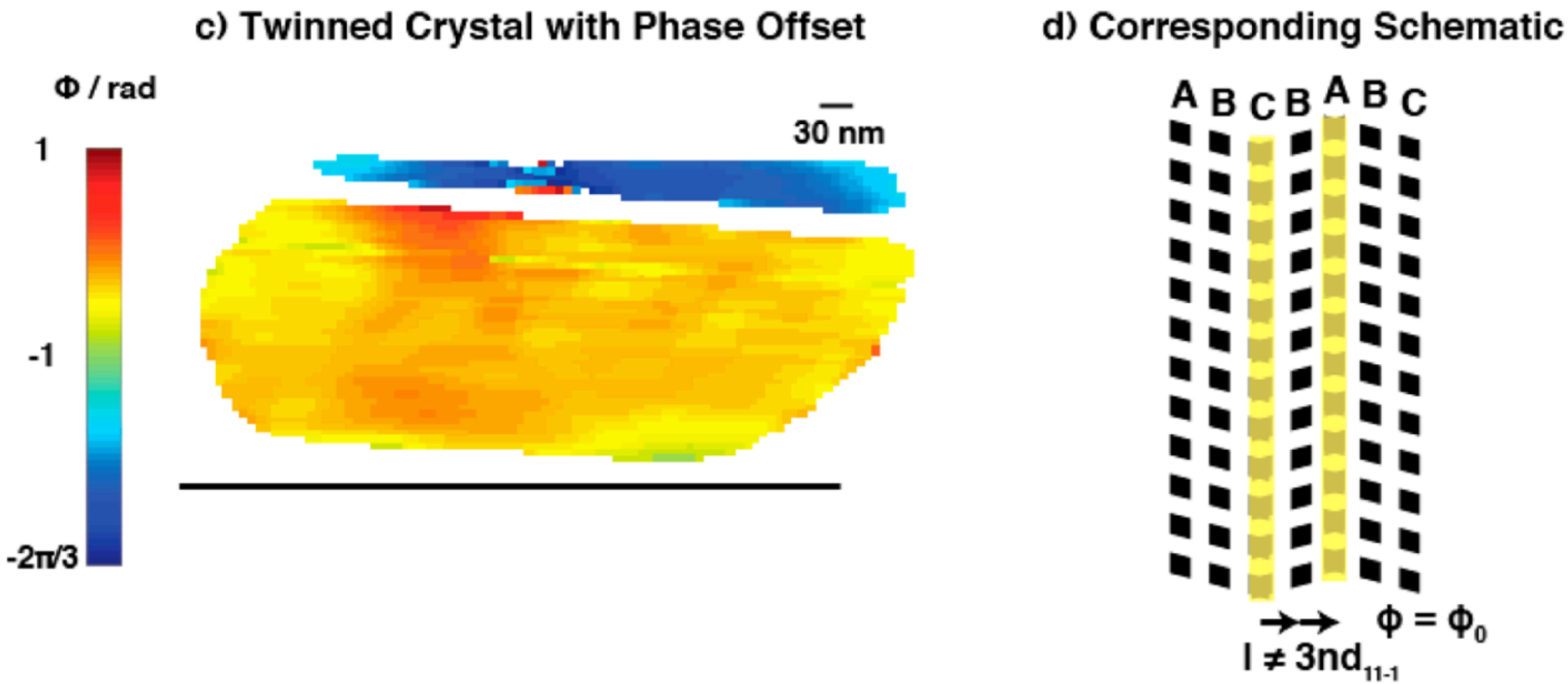


*A. Ulvestad et al. “3D Imaging of Twin Domain Defects in Gold Nanoparticles” *Nano Letters* (2015)

Defects in Real Space

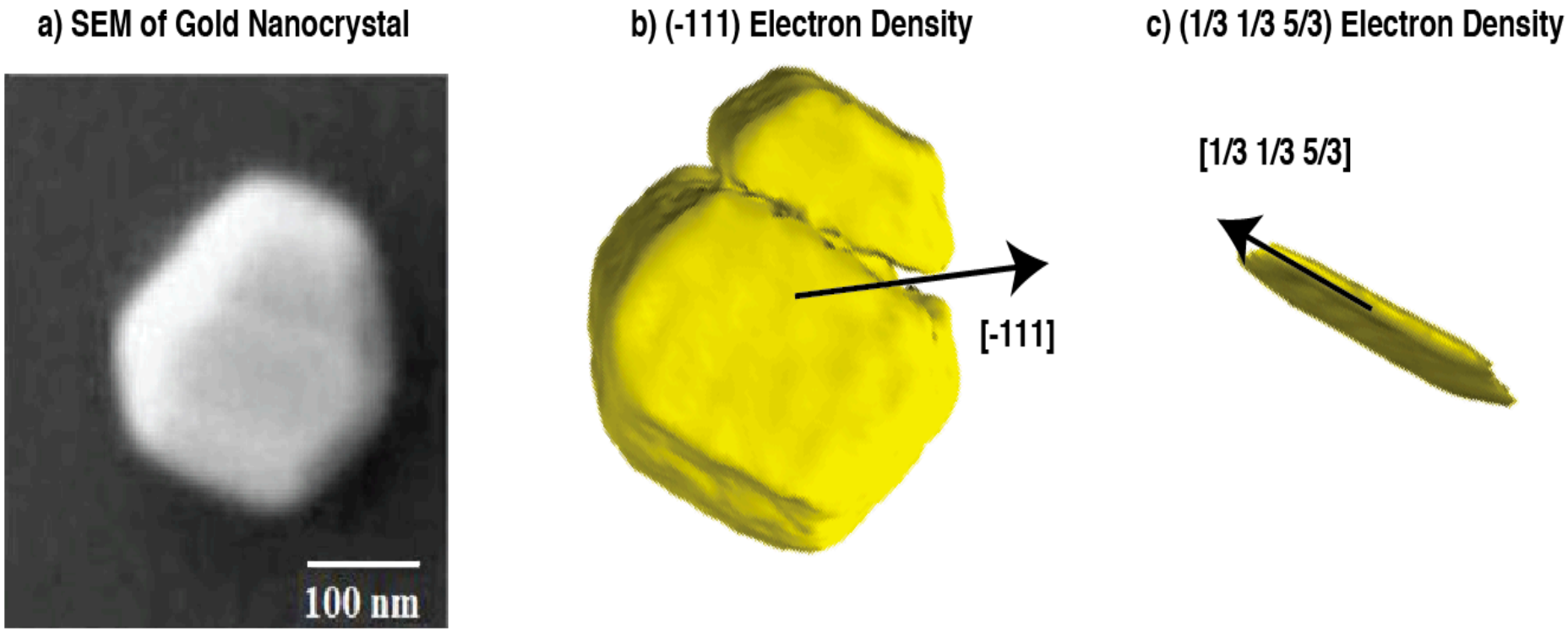


Example of Phase Offset



- Phase offset implies distance between parent crystals (twin width) not $3nd_{11\text{-}1}$
- Roughness of twin/parent interface related to standard deviation of phase in the disconnected piece

Measuring the Twin*



- Locate and measure reflection from twin planes
- Reconstructed electron density fits nicely in gap

*Measured by Ross Harder. Beitra et al. (2010)

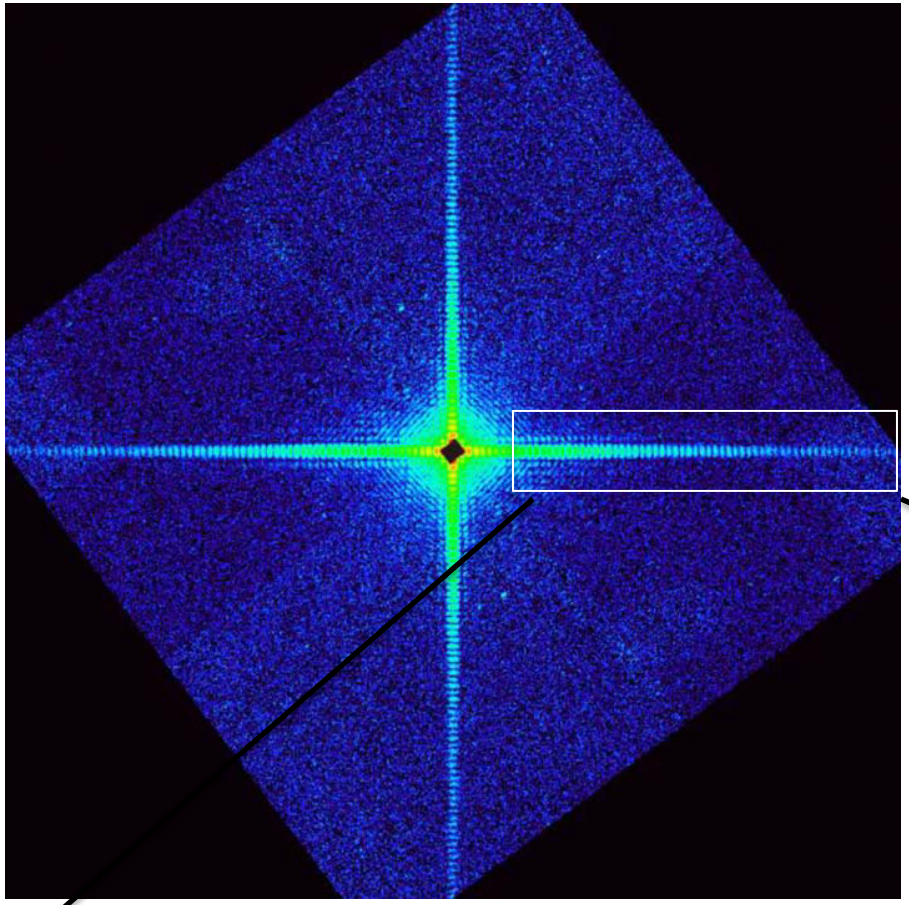
Resolution Limits?

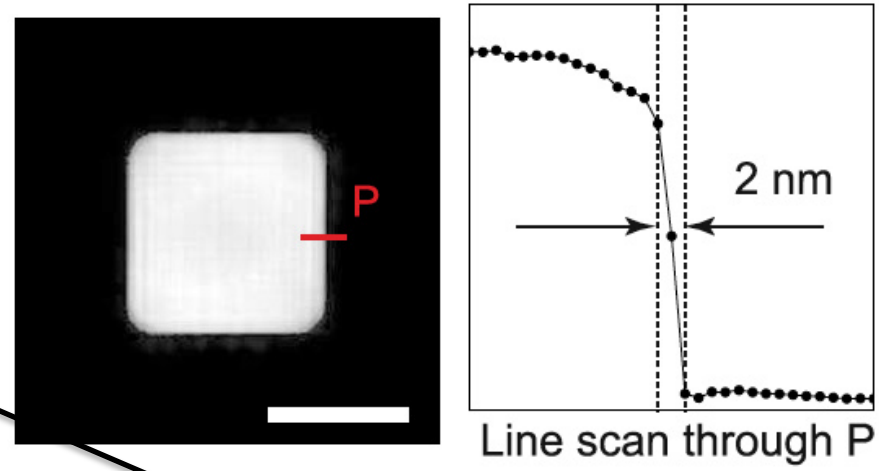
Typical resolution in CXDI is ~10-30nm

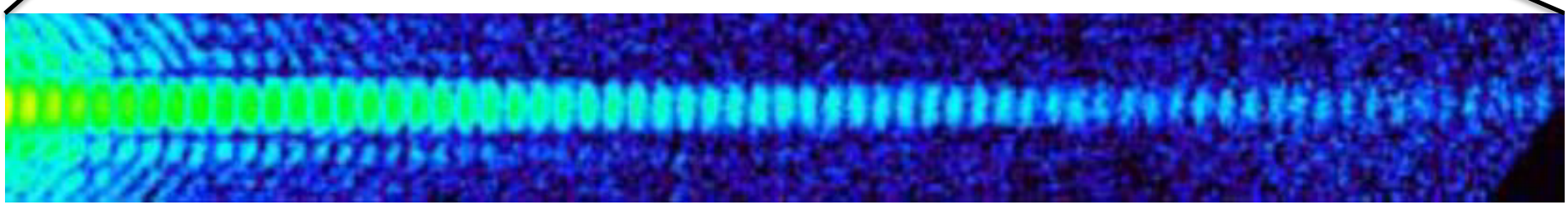
What is the ultimate limit?

Coherent Diffractive Imaging:

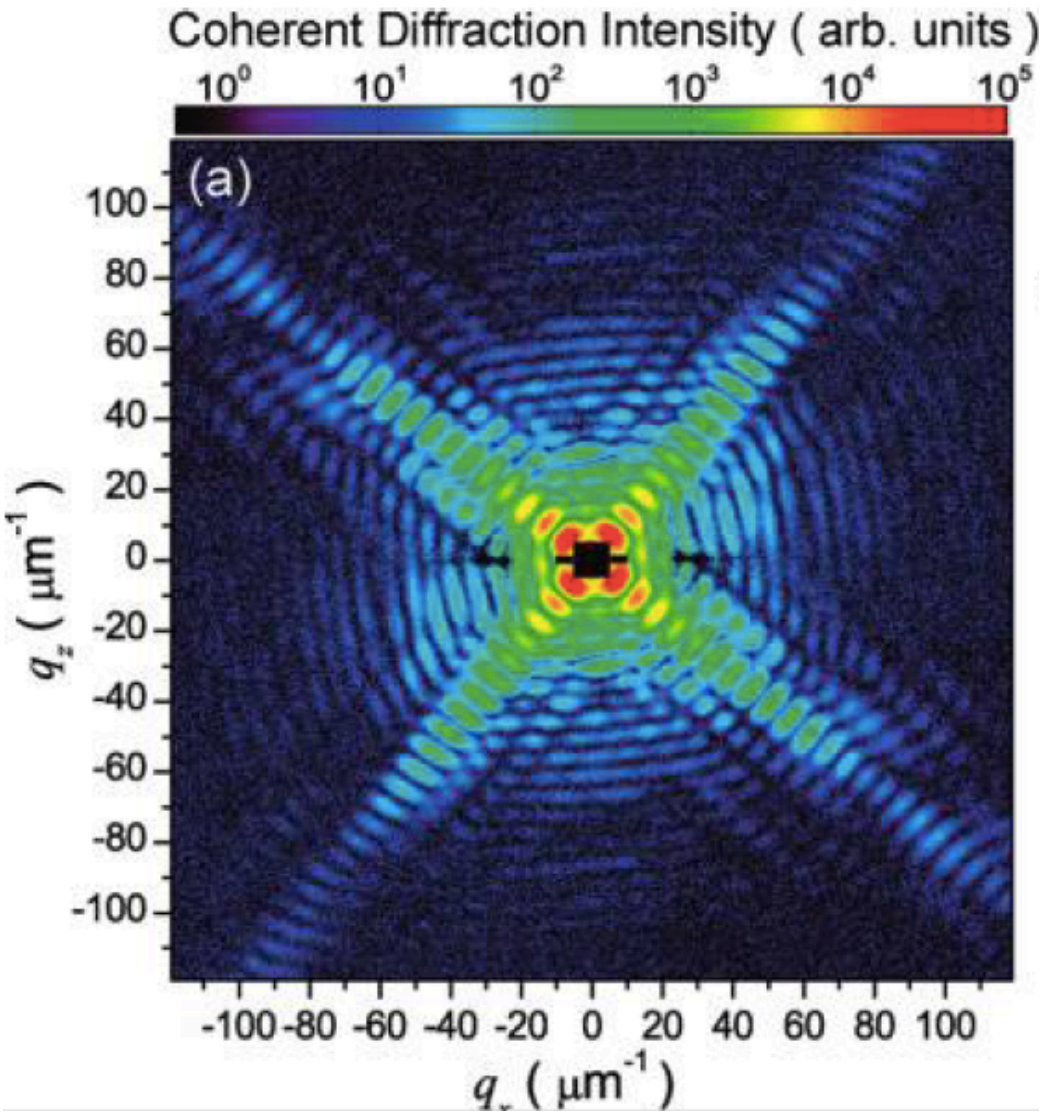
Y. Takahashi et al.,
Phys. Rev. B 82, 214102 (2010)

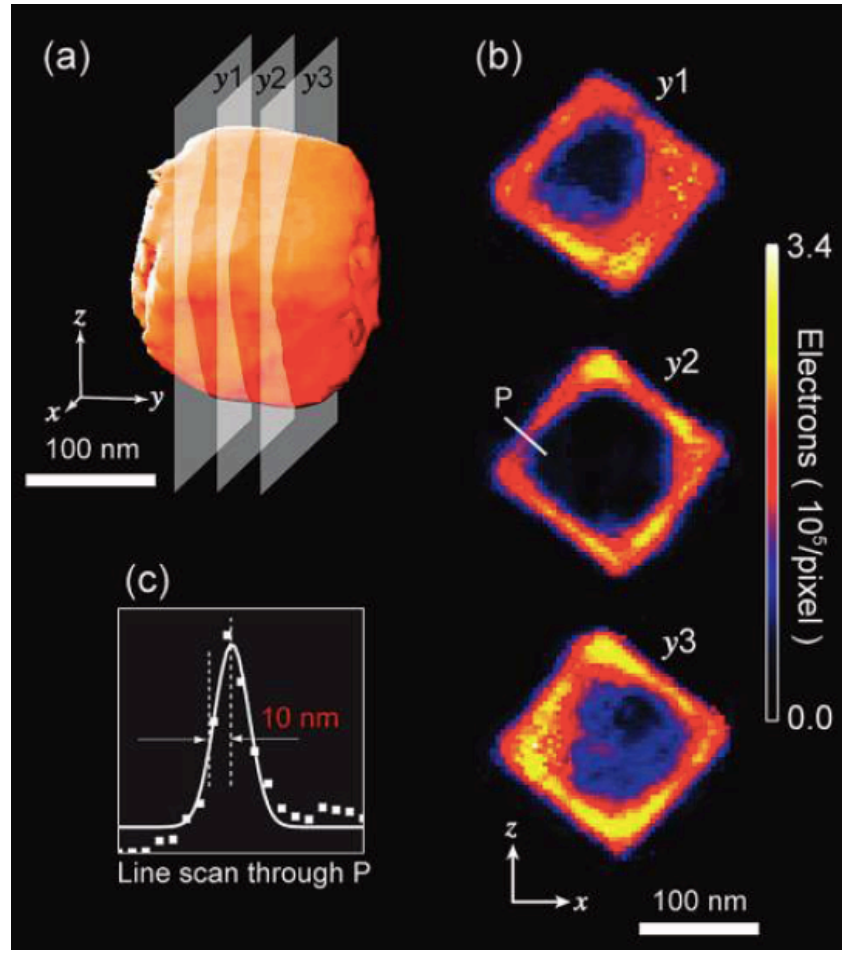






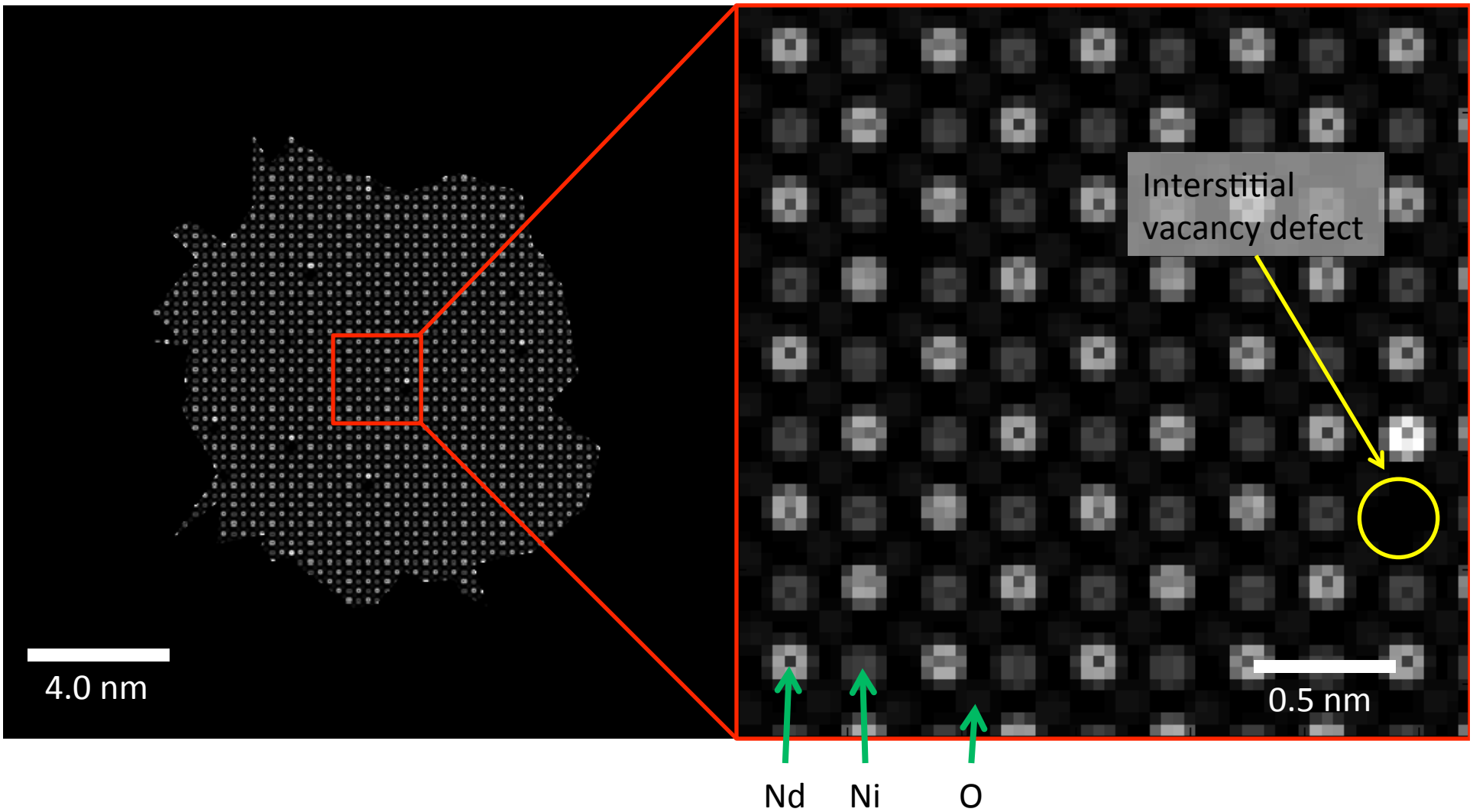
3D imaging record ~4nm





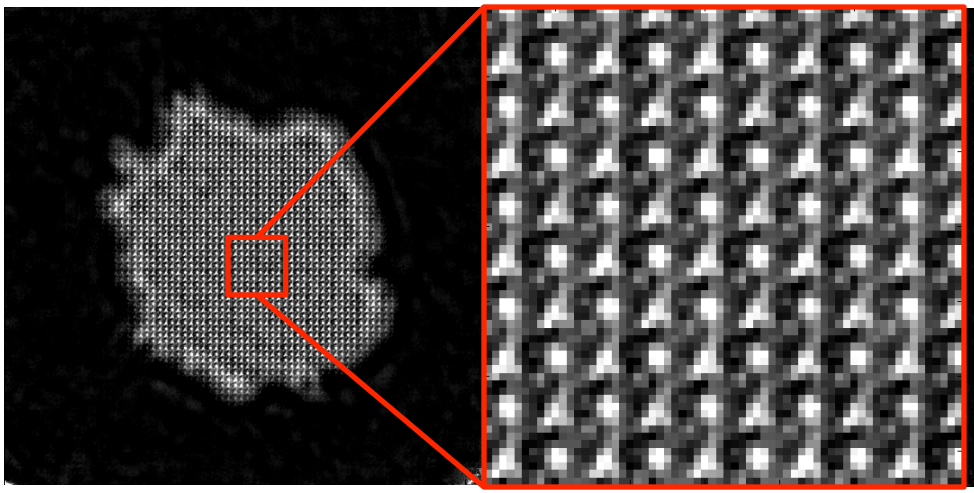
Y. Takahashi, Nano Lett., 10, 1922 (2010)

Simulatons: $NdNiO_3$ particle projected along c-axis

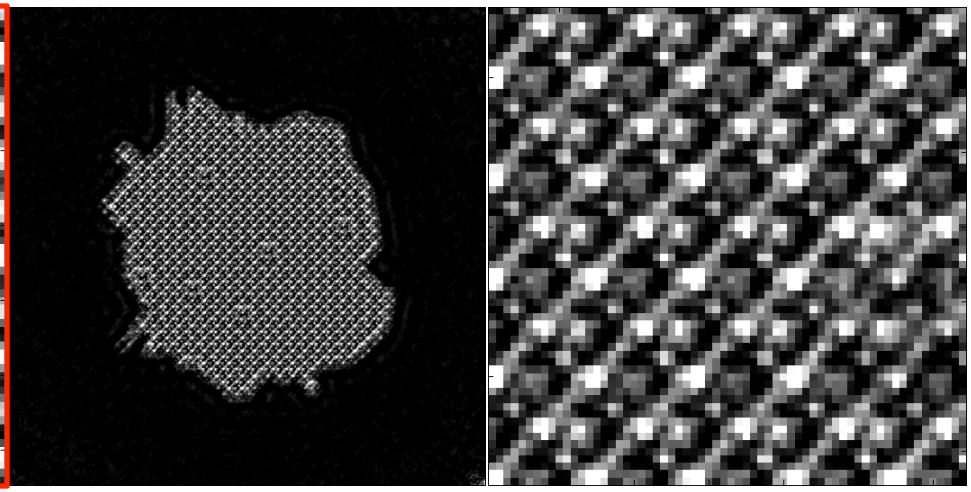


2D Reconstructions of crystalline material (with a few random, isolated defects)

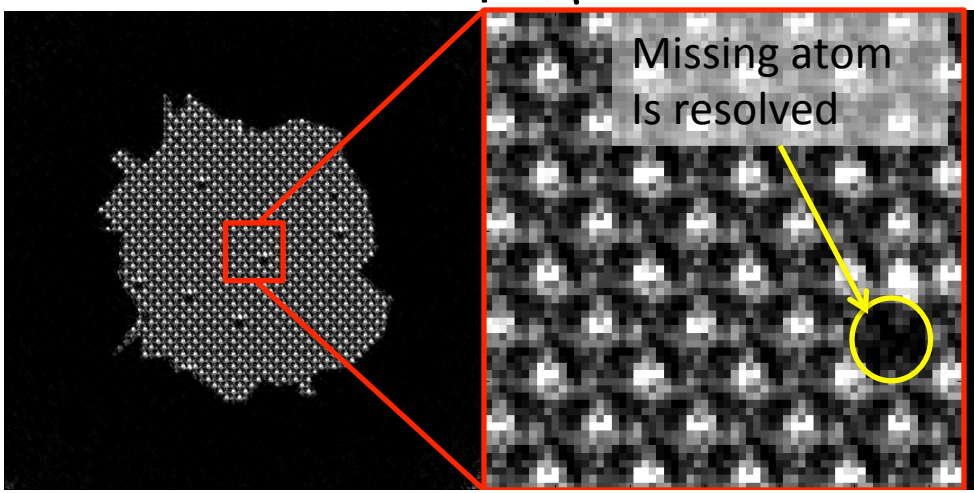
Scattered flux 1.5 x10^9 ph/μm^2



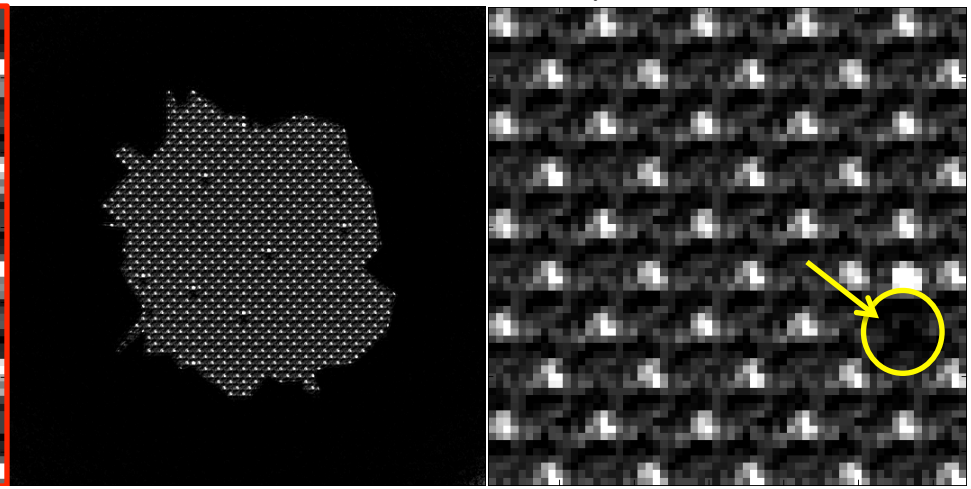
1.5 x10^{10} ph/μm^2

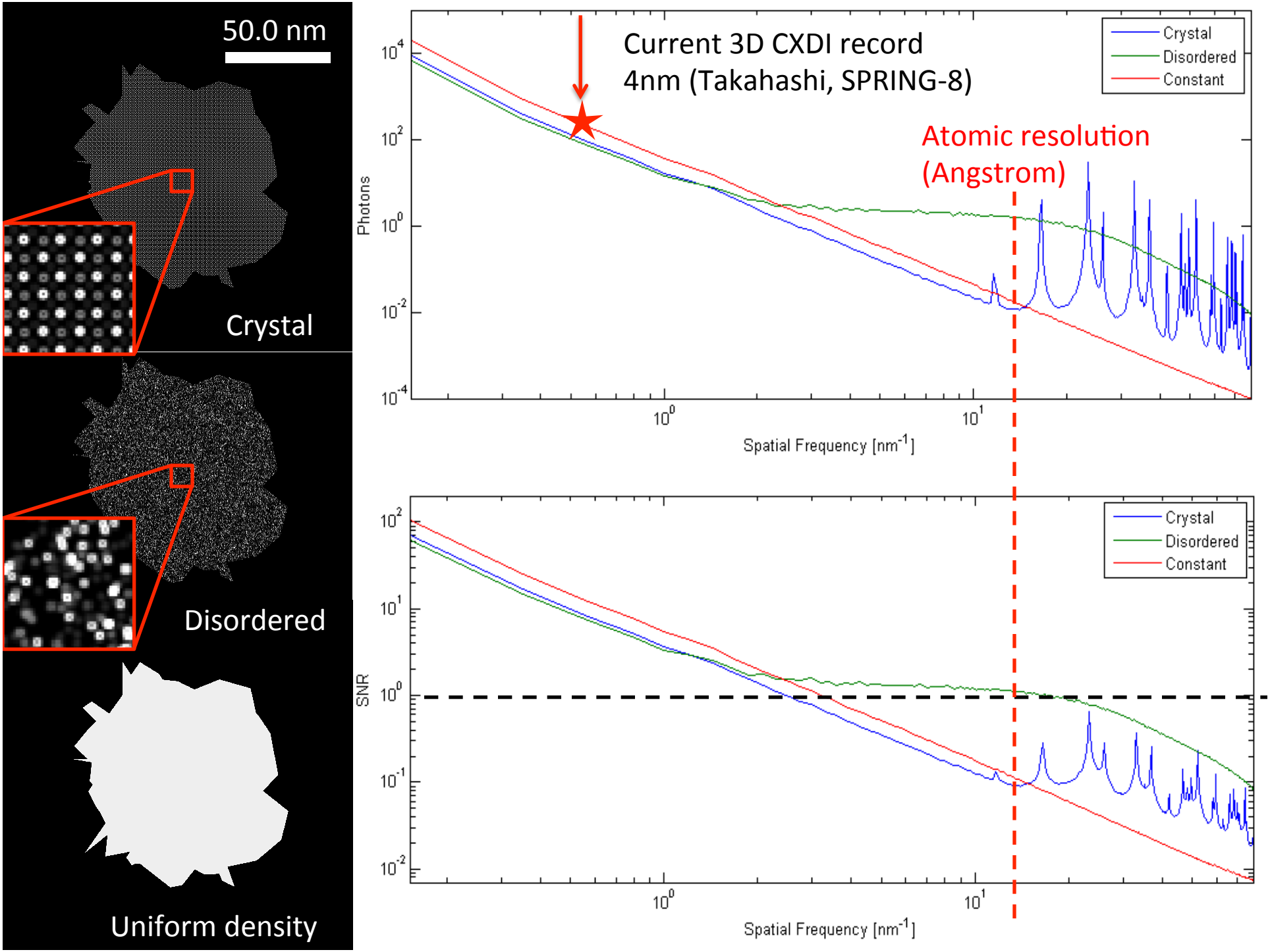


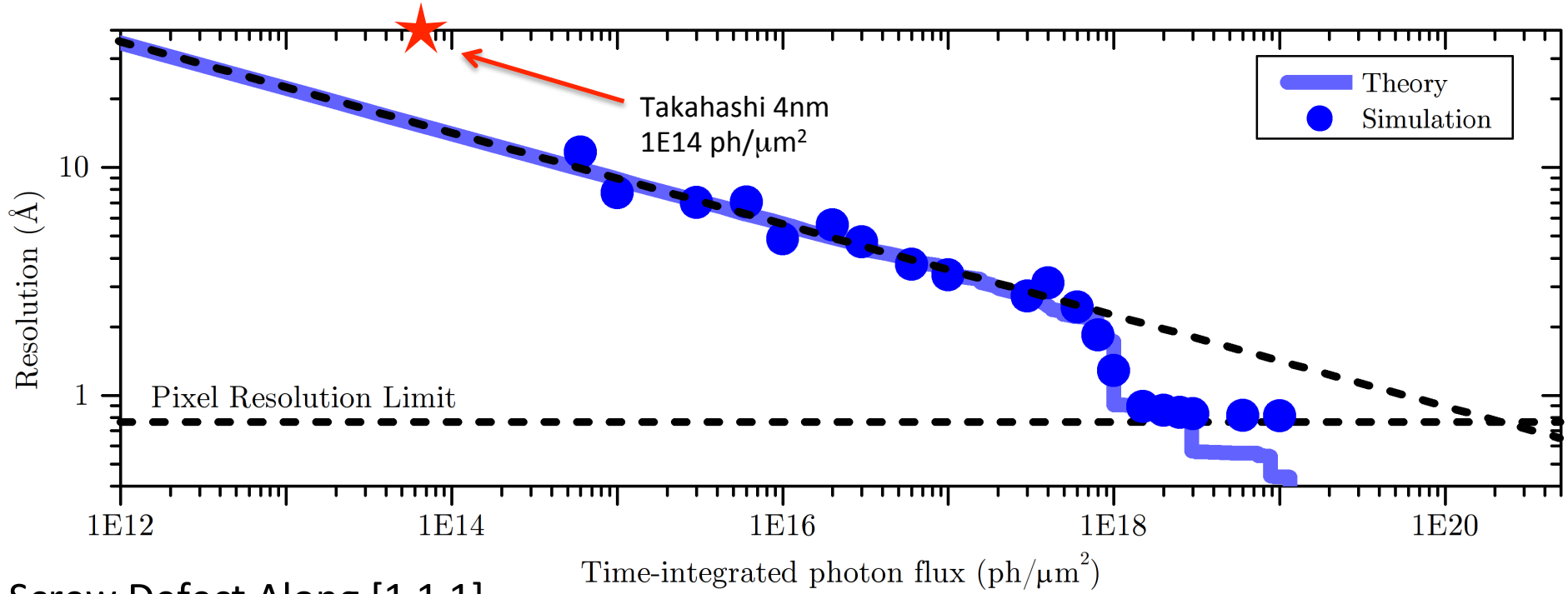
4.5 x10^{10} ph/μm^2



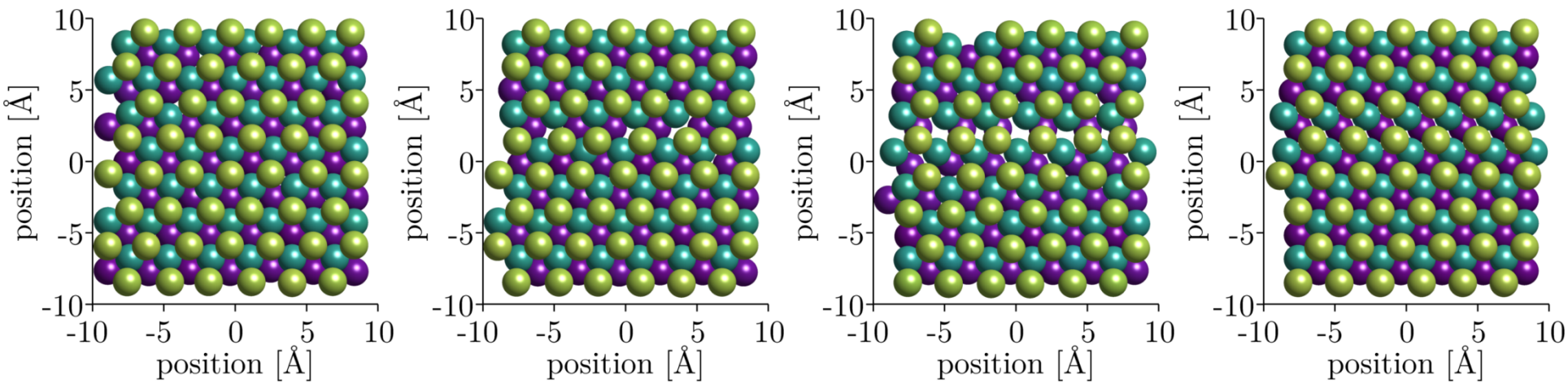
4.5 x10^{11} ph/μm^2




50.0 nm
Crystal
Disordered
Uniform density
Current 3D CXDI record
4nm (Takahashi, SPRING-8)
Atomic resolution
(Angstrom)
Crystal
Disordered
Constant
Photons
Spatial Frequency [nm^{-1}]
SNR
Spatial Frequency [nm^{-1}]



Screw Defect Along [1 1 1]

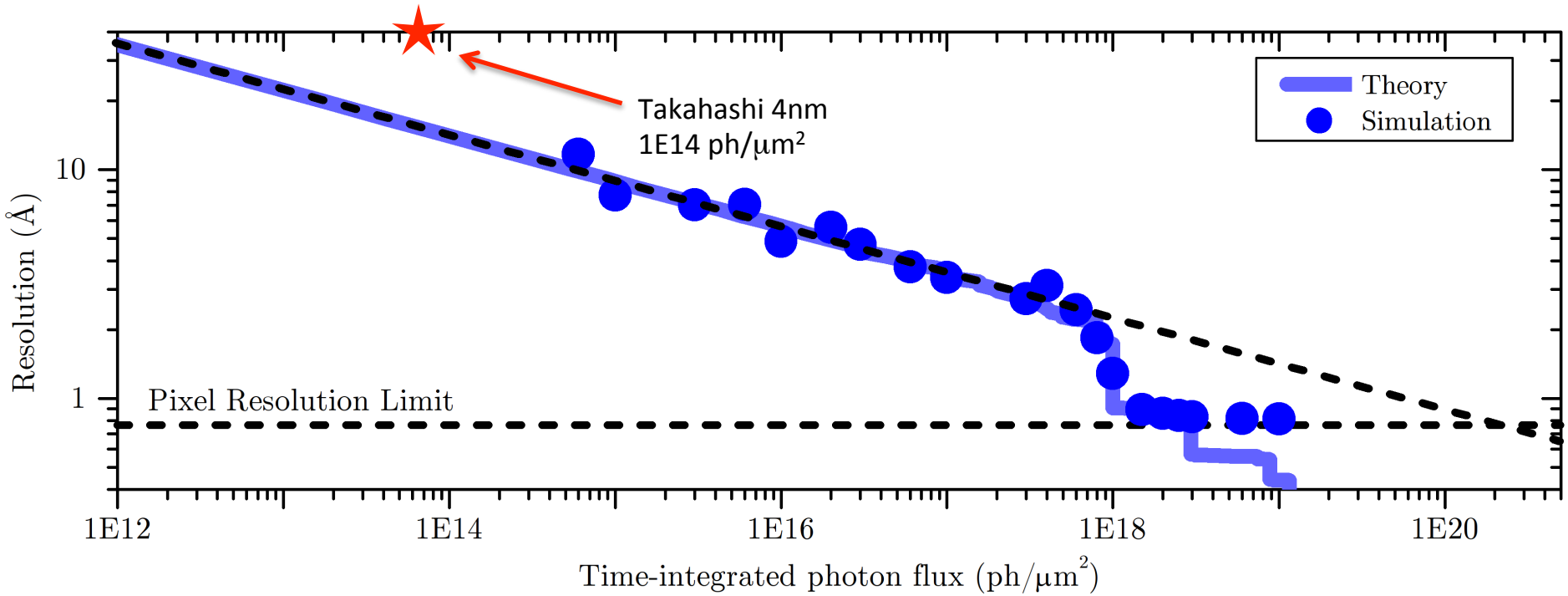


$6x10^{15}$ ph/um²
MaxE: 1.41 Å
MAE: 0.35 Å

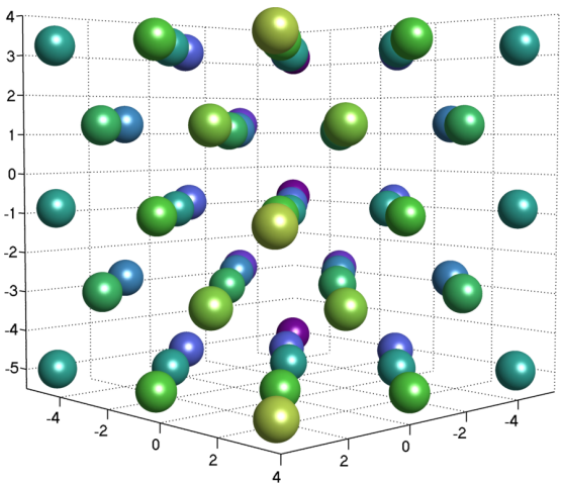
$3x10^{17}$ ph/um²
MaxE: 1.38 Å
MAE: 0.25 Å

$2.5x10^{18}$ ph/um²
MaxE: 1.07 Å
MAE: 0.25 Å

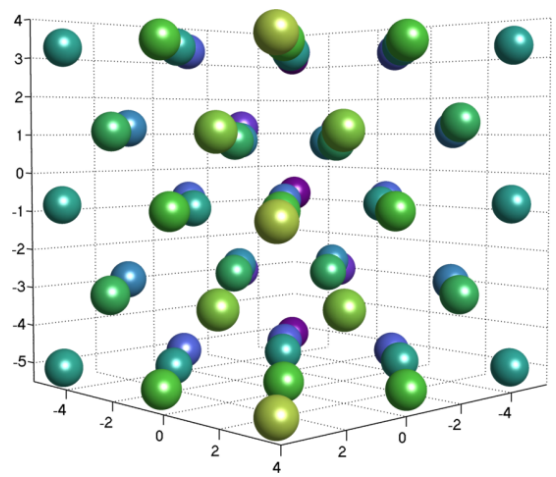
Actual



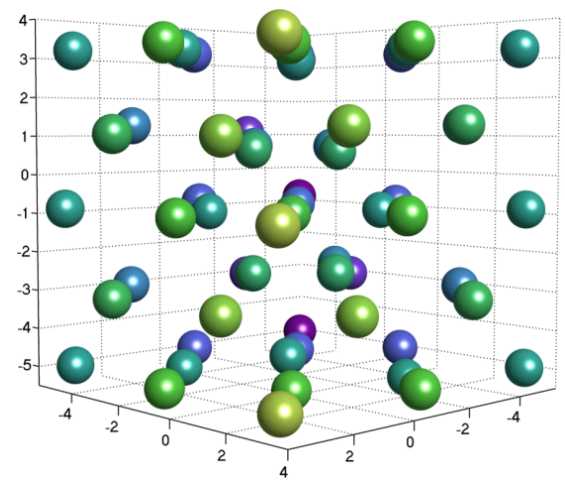
Vacancy



1.0x10^{18} ph/um^2

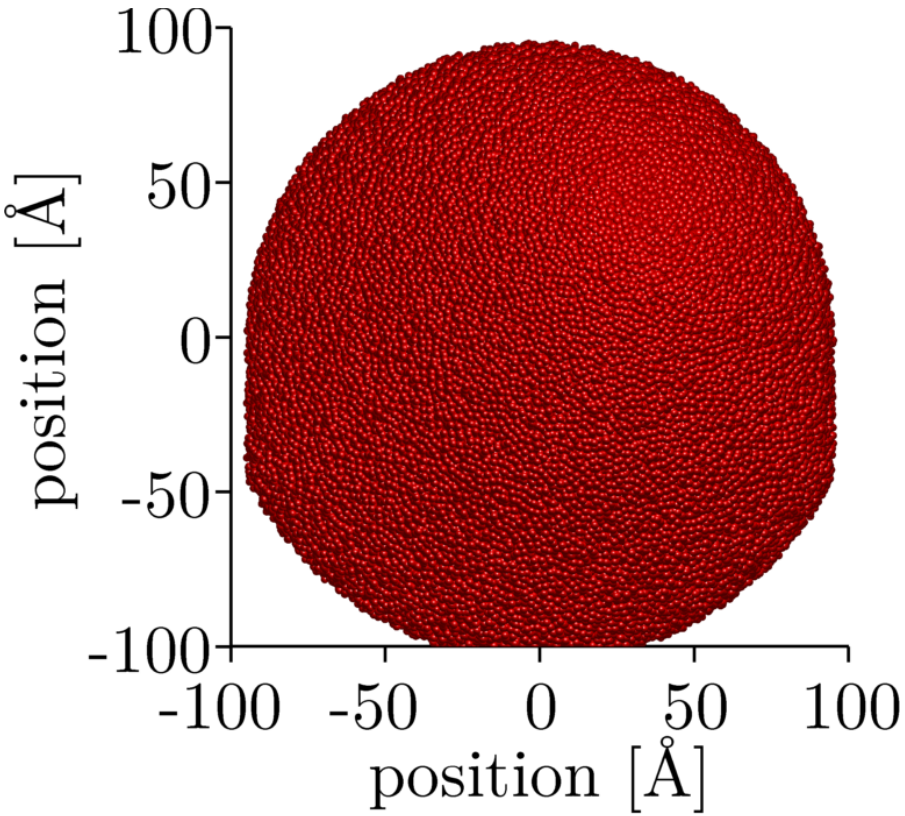


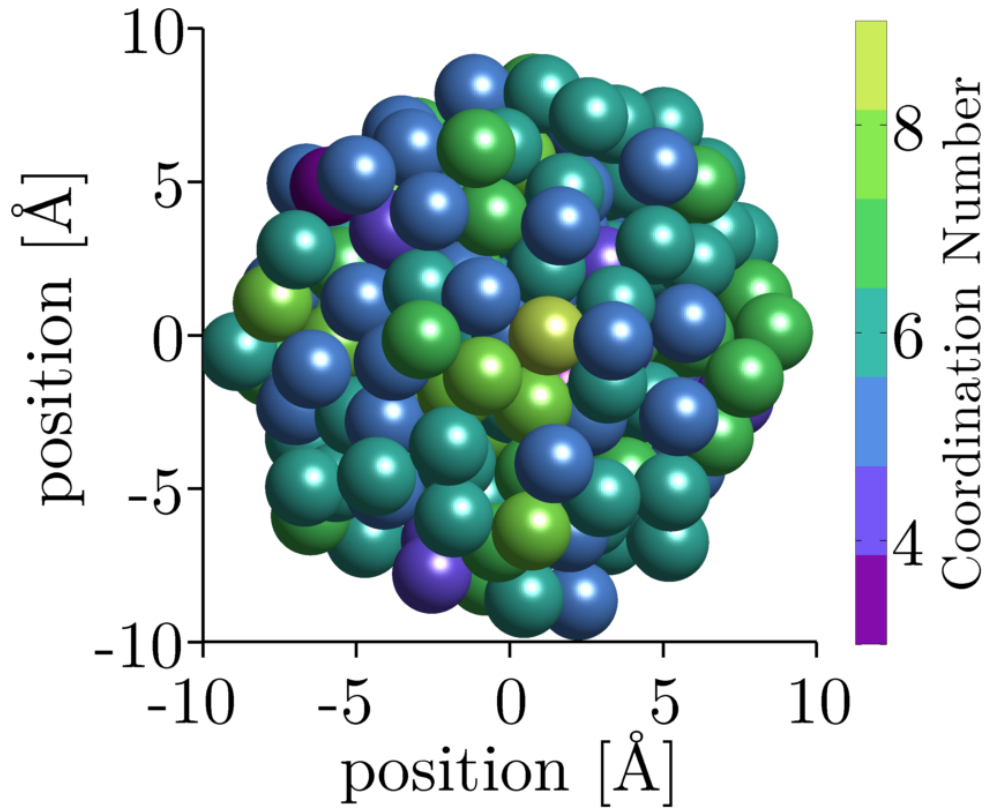
1.5x10^{18} ph/um^2

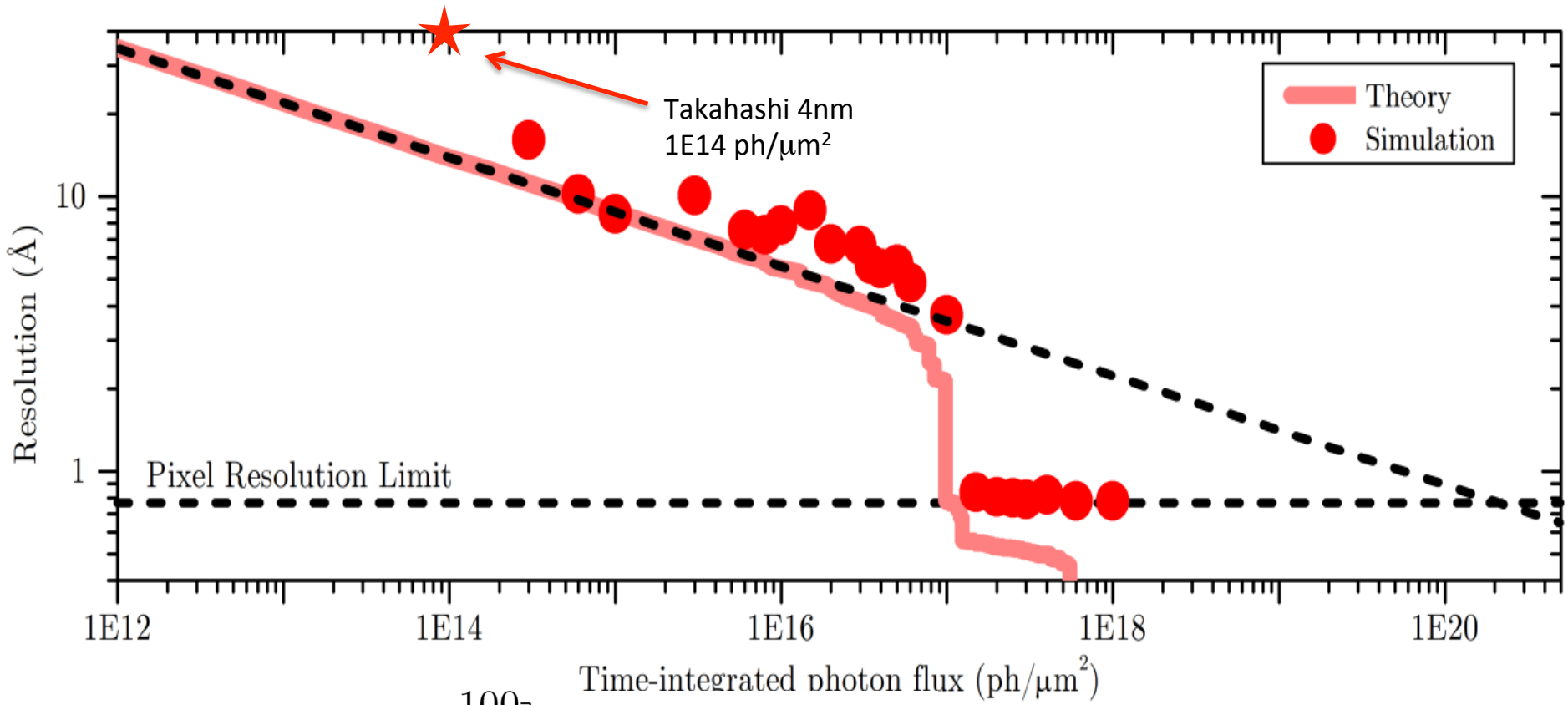


2.0x10^{18} ph/um^2

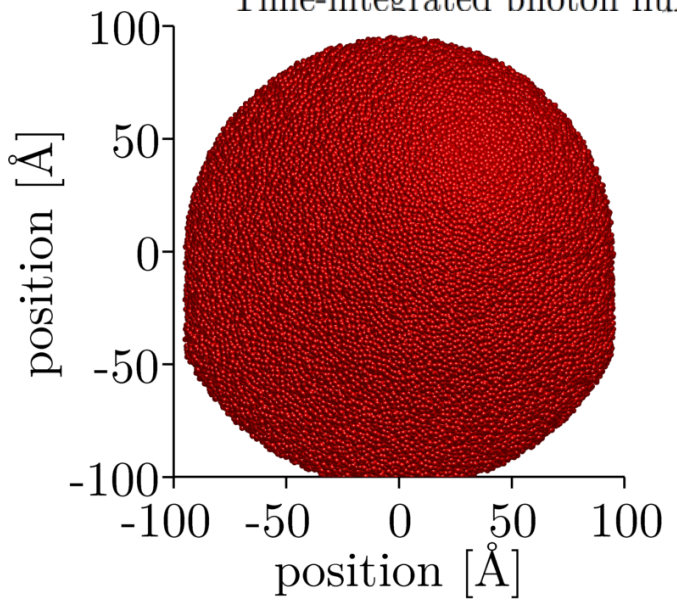
Hard sphere model for amorphous particle

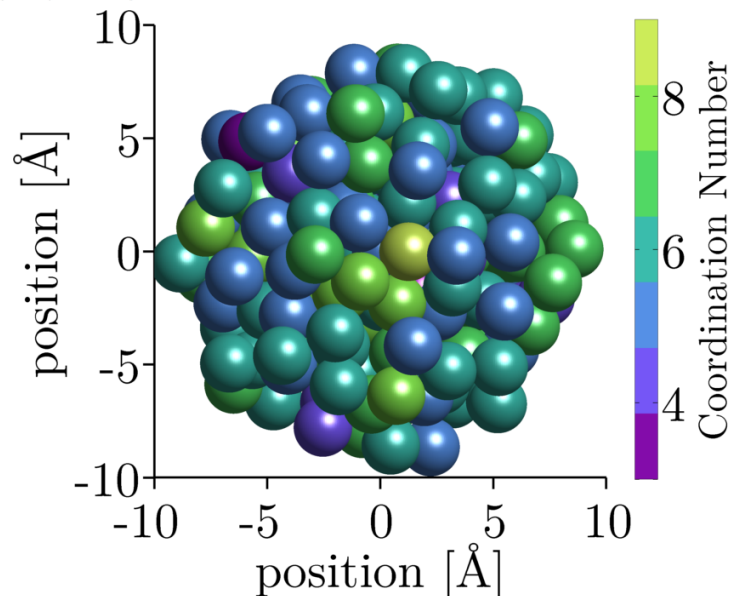






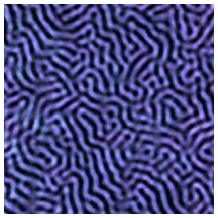
Focusing
10^{11} ph/s (APS)
Into 100nm spot
provides
10^{13} ph/s/μm²





Collaborators:

Prof. Shirley Meng and group (UCSD)



Prof. Eric Fullerton's group (UCSD)

Prof. Simon Mochrie (Yale)

- Ross Harder (Argonne)

Our Group at UC San Diego